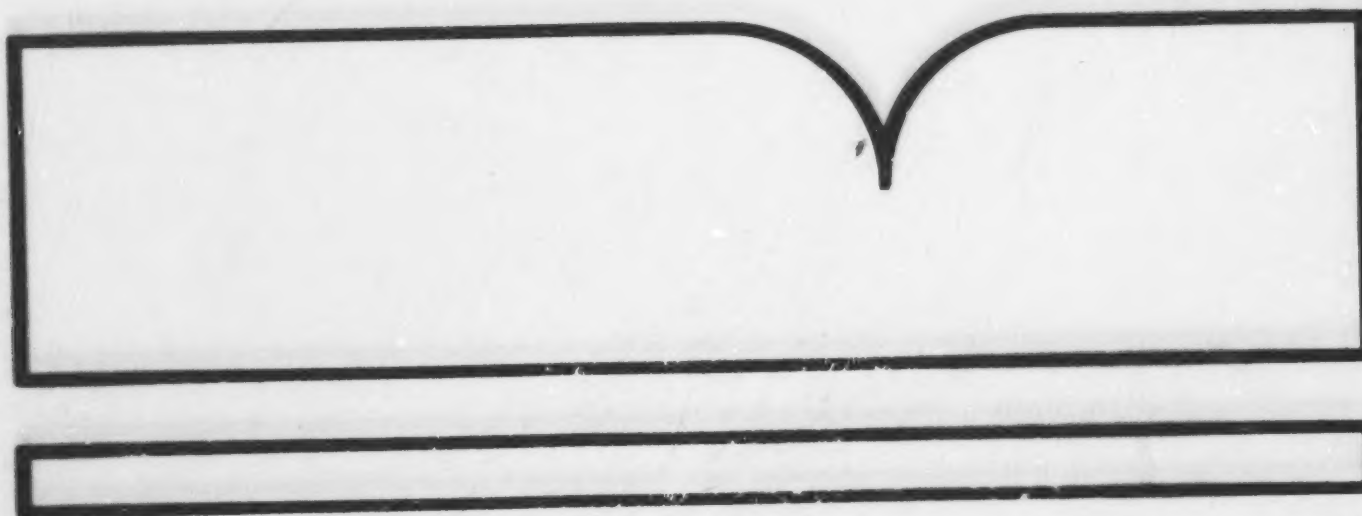


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Impact of Aerosols on Visibility and Climate
A Report Prepared for Panel 3, Environmental
Impact, of the NATO/CCMS (North Atlantic Treaty
Organization/Committee on the Challenges of
Modern Society) Air pollution Pilot Study on
Control Strategies and Impact Modeling

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IMPACT OF AEROSOLS ON VISIBILITY AND CLIMATE

A report prepared for Panel 3, Environmental Impact, of the NATO/CCMS Air
Pollution Pilot Study on Air Pollution Control Strategies and Impact Modeling

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Review comments on an earlier draft by Dr. R. J. Charlson of the University of Washington and Dr. Dale Gillette of the National Oceanic and Atmospheric Administration were a major contribution to the present document.

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I. Introduction.

This is the United States report on the impact of selected air pollutants on visibility and climate; it is a representative collection of recent research findings and key elements of data. It is hoped it meets the requirements necessary to portray the state of scientific knowledge in the United States and indicates the areas that require the policy attention of public officials.

In the selection of pollutants to be covered, the most important ones for visibility and climate (in terms of direct effects) were chosen; they include the sulfates and elemental and organic carbon. Minor contributors are mentioned only briefly. Notable exceptions are the exclusion of discussions of effects of CO₂, freons, nitrous oxide or methane on climate, which have been or will be extensively covered in other fora. Special reference is made to "Changing Climate," a report of the Carbon Dioxide Assessment Committee of the National Research Council, National Academy of Sciences (1983).

In the United States, the Environmental Protection Agency is responsible for developing air quality criteria documents covering a number of air pollutants. These documents are the result of a national attempt to collect, synthesize, and evaluate relevant research and to relate its findings to public policy and priorities. This report reflects the results of that process as manifested in Air Quality Criteria for Particulate Matter and Sulfur Oxides (U.S. Environmental Protection Agency, 1982). In addition, it incorporates several more recent research reports and the conclusions of a workshop on visibility and climate sponsored by the Environmental Protection Agency at Rougemont, North Carolina, January 29 to February 1, 1984.

II. Pollutants of Concern.

The pollutants of chief concern regarding visibility and climate are those which make up aerosols and fine particulate masses (sulfur compounds and elemental and organic carbon), those which affect the ozone layer in the stratosphere (chlorofluorocarbons, methane, and nitrous oxide), and those influencing infrared absorption (CO₂, chlorofluorocarbons, carbon, nitrogen oxides, and methane). This report will emphasize just the aerosol components.

Current knowledge indicates that fine particulate matter is composed of varying amounts of sulfate, ammonium, and nitrate ions, elemental carbon, organic carbon compounds, water, and smaller amounts of soil dust, lead compounds, and trace species. Sulfate often dominates the fine mass and light scattering,

while elemental C is sometimes an important visibility-reducing species in urban areas. Significant variations can occur at different times and sites. The 30-year record of the spatial and temporal trends of coal combustion and visibility suggests that the increasing emissions of SO_x since the 1950's have been associated with similar increases in haziness. Nevertheless, the relationship between SO_2 emissions and resulting sulfate concentrations is not as well known as the relationships between sulfate concentrations and visibility reduction.

There is little record of SO_2 and sulfate concentrations before 1970, particularly in non-urban areas, but some data have been compiled by Altshuller (1980). Between 1963-65 and 1971-73, the three-year average northeastern U.S. urban SO_2 concentrations decreased by 80 percent, reflecting the decreases in urban sources required by the implementation of the Clean Air Act. Further decreases of about 15 percent occurred during the 1970's at the urban sites. Measurements of SO_2 at non-urban sites are often near or below the levels of detectability. Annual average concentrations are around $10 \mu\text{g}/\text{m}^3$ and fourth quarter concentrations are usually higher than the other quarters. Altshuller could find no observable trend in non-urban SO_2 concentrations.

Sulfate measurements in the past were subject to errors due to the production of "artifact sulfate" produced by the oxidation of SO_2 to sulfate on the filters used to collect the aerosol, and Altshuller assumes this error is small in his analysis. More recent sulfate collection techniques do not have such a substantial error.

Three-year running averages of annual average sulfate concentrations for New York State are presented in Figure 1. The range of Altshuller's data for the second, third, and fourth quarters of the year is also shown as the shaded area. The figure shows that New York's sulfate data, which were obtained from a mixture of non-urban and urban sites, fall within the range of Altshuller's non-urban values for the Northeast. Rochester, which could be classified as an urban site, represents the longest record at one station, and is also shown to be in the range of Altshuller's data. The graph shows that there has been a modest decrease (about 20 percent) in New York sulfate concentrations during this period. This decrease is primarily due to decreases in urban stations. The Rochester station shows no change over this 17-year period.

Altshuller's data show little change in sulfate concentrations in non-urban sites in the Northeast and decrease in sulfate concentrations at urban

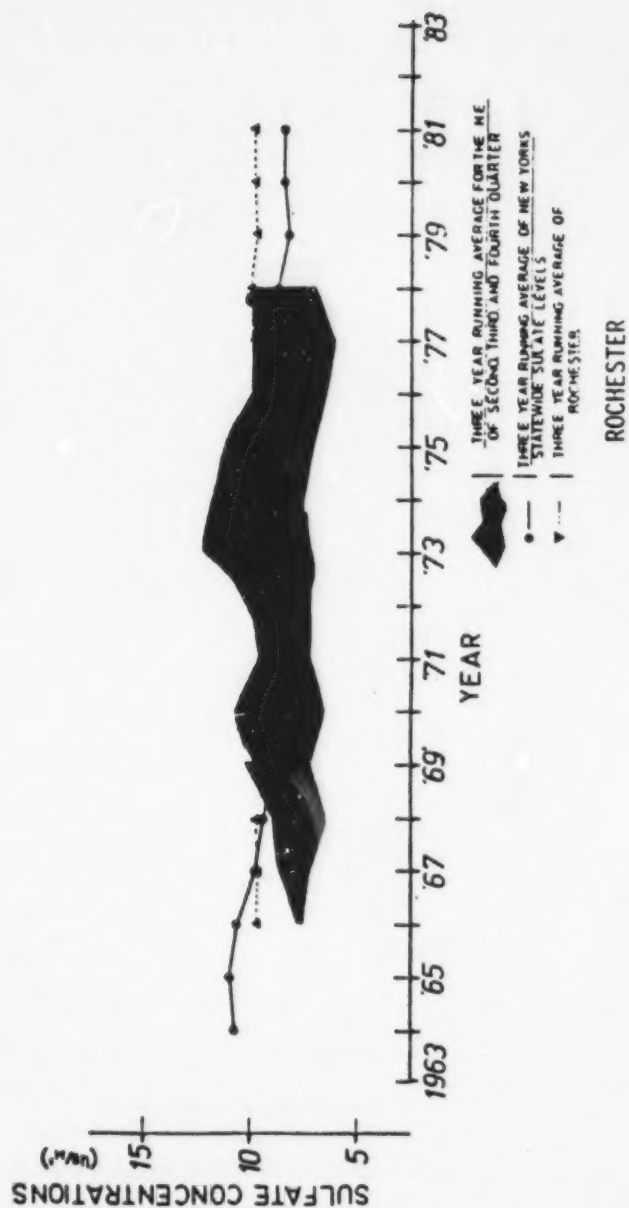


Figure 1. Sulfate concentrations in the northeastern United States.

sites. The New York State data show similar trends. SO_2 has declined at all sites, and it can be concluded that there have been no general increases in concentrations of either SO_2 or sulfate in the Northeast as a whole over the past 20 years.

Several studies have shown that sulfate constitutes 40 to 50 percent of the fine particle mass in the Eastern U.S. and 30 to 40 percent in the West. It is dominated by compounds with composition ranging between H_2SO_4 and $(\text{NH}_4)_2\text{SO}_4$ (Charlson et al., 1974).

Further studies of the humidity dependence of light scattering demonstrated that these sulfate aerosols were not limited to urban settings but were distributed over a region of at least 100/km extent (Weiss et al., 1977). Sulfate aerosol mass concentration was determined along with the molecular characterization by response to humidity showing that light scattering by particles is not only well correlated with sub μm mass, but with sulfate concentration as well (Waggoner et al., 1976).

The second most frequent components of aerosol and fine particulate masses in the U.S. are organic and elemental carbon. While sulfate often dominates the mass and light scattering, elemental carbon from soot is commonly the primary light-absorbing matter in urban areas. See Figures 2 and 3 for examples of particle light extinction (particle scattering and particle absorption) apportioned by chemical species for cities in Texas and California.

Means have been developed to measure the light absorption coefficient due to particles (see Lin et al., 1973, and Clark, 1973). Simultaneous measurement of light scattering and absorption showed that the eastern U.S. was also influenced by persistent absorption due to elemental carbon C(0). The combination of these two measurements yielded a direct determination of the albedo of single scatter, w , which has values around 0.9 in the eastern U.S., down to as low as 0.5 in some cities (Charlson and Robinson, 1984; Waggoner et al., 1981).

Sources of atmospheric particulate matter are both anthropogenic and natural. Anthropogenic sources include combustion of petroleum, coal, and wood, while natural sources include dust storms, volcanic emissions, forest fires, and windblown sea salt. While forest burning has decreased dramatically since 1900, industry and internal combustion engine sources have increased dramatically.

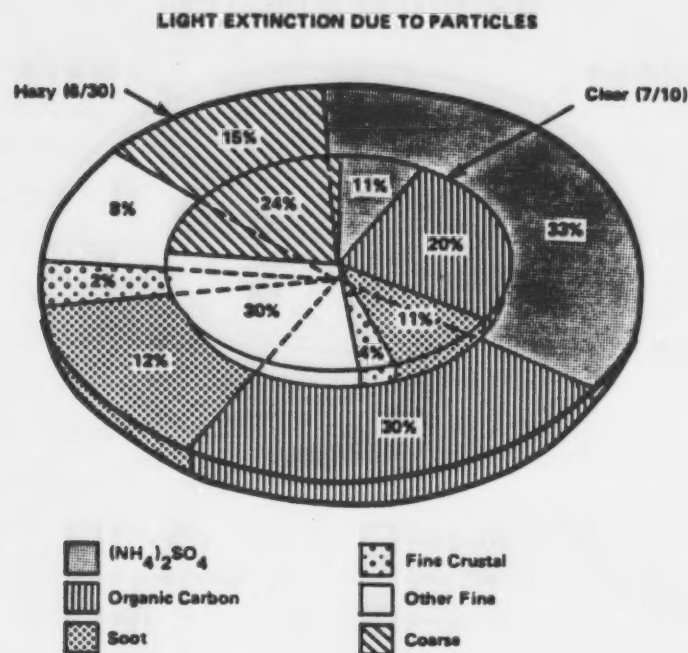


Figure 2. Particle light extinction ($\sigma_{sp} + \sigma_{ap}$) budget for the low visibility southern California incursion (June³⁰) and a clear day (July 10). On the hazy day ammonium sulfate and organic carbon accounted for 63% of the particle light extinction, while on the clear day these species accounted for only 31%.

Source: Macias et al. (1981a,b).

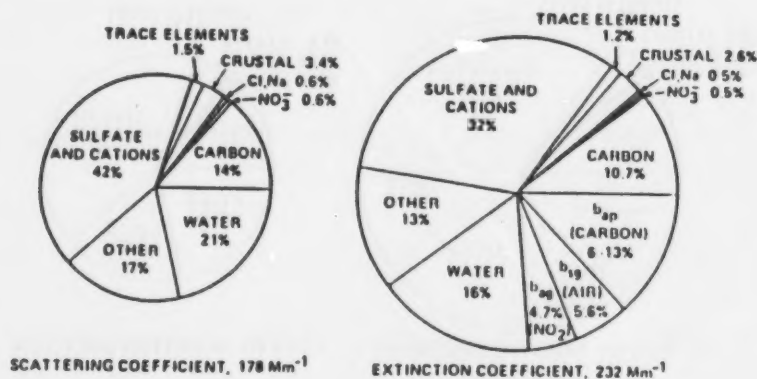


Figure 3. Eight-day average of daytime light scattering and extinction coefficients apportioned by chemical species in Houston from September 11 to 19, 1980.

Source: Dzubay et al. (1982).

In addition to these direct sources, there is also secondary production by conversion of reactive gases to particles. After their introduction into the atmosphere, particles interact with each other and with atmospheric gases, and are ultimately removed to the earth's surface. The main gaseous precursor of particles is SO_2 , which is primarily anthropogenic.

In the U.S. the following trends have been observed or may be predicted and are relevant to the following discussion:

1. Regional sulfur oxide emissions will go up through 1995, thereby leading to further decrease in visibility in some locations.
2. Particulate matter emissions from power plants will go down significantly within the next 30-50 years due to new-source emission standards.
3. If current acid rain strategies are put into effect, visibility should improve strikingly.
4. Wood smoke is on the increase, but the question is, does domestic burning equal forest burning? Smoke levels indicate the problem is regional.
5. NO_x will increase over the long term.
6. Hydrocarbons will go down with cleaner combustion in automobiles.

III. Impact on Visibility.

In the 1970's, interest in pollutant effects on visibility increased because of the "environmental movement." Deterioration of visual air quality due to manmade aerosols was studied extensively, especially after the passage of the Clean Air Amendments of 1977 (section 169A) stating that the scenic beauty of national parks and wilderness areas should be protected.

"Atmospheric visibility" is a term used by all airport weather observers to connote visual range, which refers to the farthest distance at which a large, black object can be seen against the horizon sky in the daytime. In the everyday sense, visibility relates to atmospheric clarity and the perceived characteristics of viewed surroundings, including the contrast and the color of objects and sky.

Traditionally, visibility has been defined in terms of the distance from an object that is necessary to produce a minimum detectable contrast between that object and its background. The type and degree of impairment are determined by the distribution, concentrations, and characteristics of atmospheric

particles and gases, which scatter and absorb light traveling through the atmosphere. Scattering and absorption determine light extinction.

The currently available methods for monitoring visibility measure different aspects of visibility impairment. Generally, contrast-type measurements (such as photography, telephotometry, and human eye observations) relate well to the perception of visual air quality, while extinction or scattering measurements (such as transmissometry and nephelometry) relate to the cause of visibility degradation. Each of these measurement methods can be used to approximate visual range.

Surface observations by human observers are reported to and disseminated by the World Meteorological Organization. The data set includes about 5000 stations over land areas but not the data for the marine reports from ships. A global network of about 100 stations also reports the daily aerosol vertical optical depth, as turbidity, when the sun is unobscured by clouds.

In the 1970's it was recognized that the synoptic network of visual range observations provides an attractive data base for study of the evolution, transport, and dissipation of synoptic scale hazy air masses (Hall et al., 1973).

On a global scale, the visibility and turbidity data have severe limitations. Prior to any statistical or other analysis, it is essential to scrutinize the data. Two chief problems are incomplete records and inconsistent use of visual targets.

A systematic bias of visual range data at a given station arises from the fact that the targets are in uneven intervals and there is an upper limit of visual range beyond which the data are not resolved. In remote flat terrain the maximum visual range reported may be as low as 10 kilometers, in which case most data are above this threshold.

Variability in the time of day and relative humidity when observations are made can also limit the utility of visual range data. Daytime visual range reduction is due to the solar radiation scattered into the visual path by the aerosol as well as by light extinction. At night the visual deterioration is due to the light extinction only. Thus daytime visual range depends primarily on aerosol extinction coefficient while at night the visual range depends on the source intensity as well. The diurnal pattern of light extinction coefficient calculated for fixed relative humidity (RH) increments is shown in Figure 4 for Dayton, Ohio. It is apparent that there is about a

factor of two increase of daytime extinction coefficient compared to the night values. This is attributed primarily to the incompatibility of visual range determinations at day and night. When visual range is used for the estimation of the aerosol extinction coefficient, only the daytime values should be analyzed.

The thirty-year trend of quarterly eastern U.S. extinction coefficient is shown in Figure 5. During the first quarter of the year, the noon mean extinction coefficient, taking all observations, was about 0.55 km^{-1} and it has been roughly constant since 1948. Once the observations coinciding with fog or precipitation were ignored, the mean b_{ext} dropped to about 0.3 km^{-1} , constant over the years. The RH correction yielded a further drop to about 0.25 km^{-1} . The trends in the fourth quarter were similar to those in the first quarter. The most interesting trends are shown in the third quarter. The exclusion of precipitation events decreased b_{ext} only about 10-20%. The RH correction to the b_{ext} was insignificant. The trend of the summer haziness shows that in the early 1950's, the RH corrected b_{ext} over the eastern U.S. was about 0.2 km^{-1} , rose rapidly to 0.3 in the 1960's, and dropped below 0.3 in the 1980's.

Once the percentiles for each station are established, other properties of the distribution function can be calculated, e.g. the total dosage, the episodicity (what fraction of the dosage is contributed by the worst 10 or 20% of the values), etc. For example, one can calculate the difference of two maps, b_{ext} for 1976-80 and b_{ext} for 1950-54 (Figure 6). The maps of b_{ext} increase for quarters 1 and 3 are shown in Figure 7. Evidently in the winter quarter the haziness has increased in the southeastern and northwestern states. The increase of summer haziness occurred mostly in the southeast, east of the Mississippi and south of the Ohio rivers.

Meteorological parameters, e.g. temperature, humidity, wind direction, wind speed, solar radiation, etc., influence visibility through dispersion of aerosols, or by changing their properties or formation and removal rate. Temperature and relative humidity may also indirectly influence the emission rate of aerosols or the precursor gases, e.g. for electric utility plants at high summer temperature and relative humidity. The humidity classified extinction coefficients (Figure 8) show that the average haziness increases with humidity. This, however, does not necessarily imply that relative humidity is the cause of high extinction coefficients. In order to eliminate relative humidity as a variable, it is advantageous to perform the trend analysis using only data for fixed relative humidity windows.

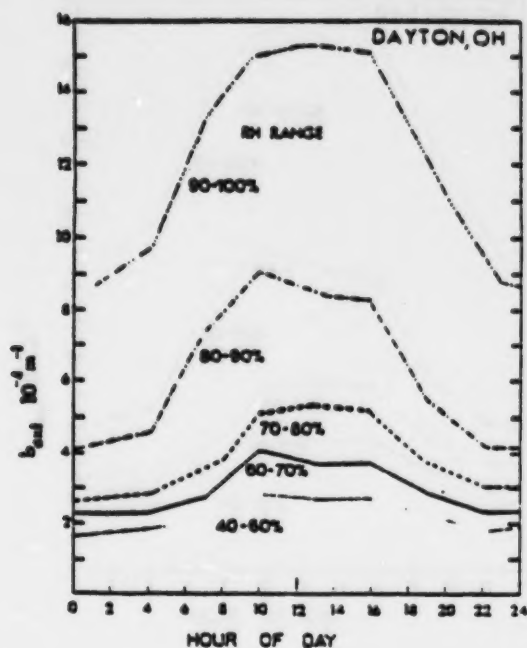


Figure 4. Visibility data, classified according to time of day and relative humidity ranges, Dayton, OH, 1970-74 (Husar et al., 1979).

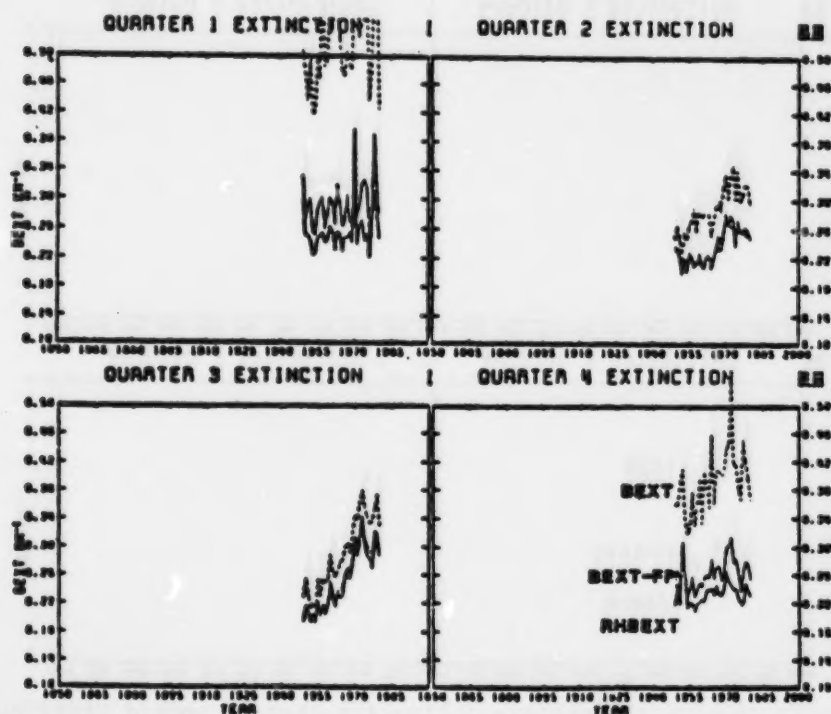


Figure 5. Quarterly trends of mean extinction coefficient over the eastern U.S. The top dashed line reflects all values, the middle dotted line only those values exclusive of fog or precipitation events, and the bottom solid line those values exclusive of fog or precipitation events corrected for relative humidity.

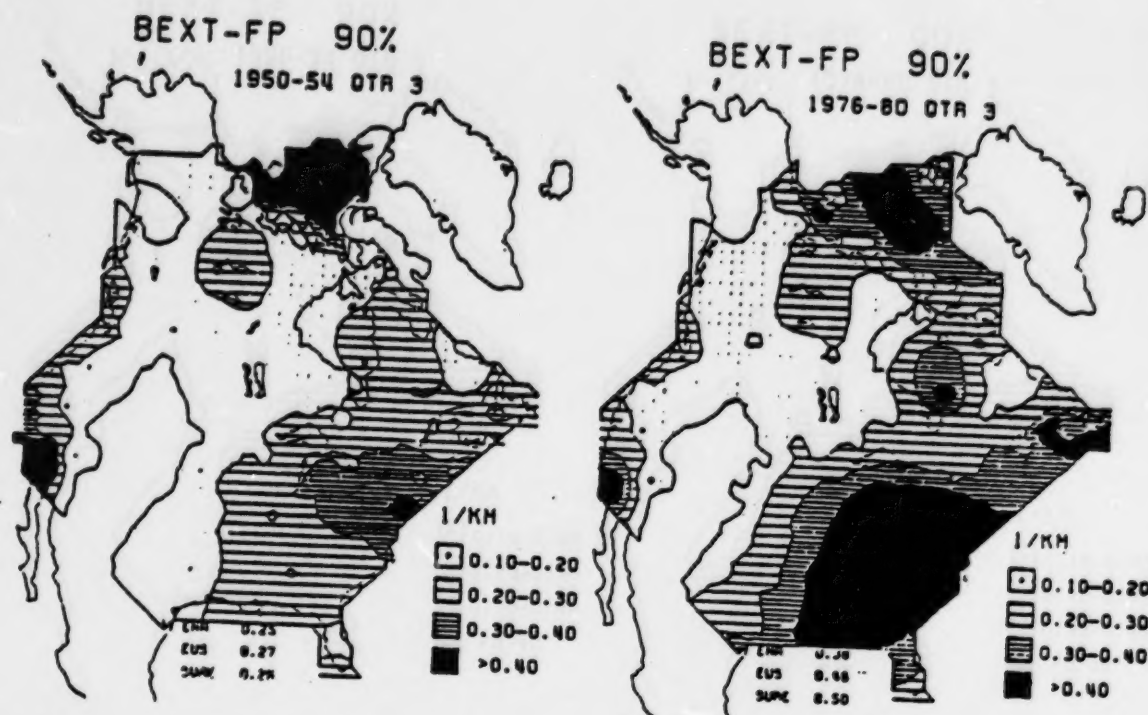


Figure 6. Spatial distribution of 90th percentile extinction coefficient for July, August, and September, 1976-1980. All values excluding fog or precipitation events.

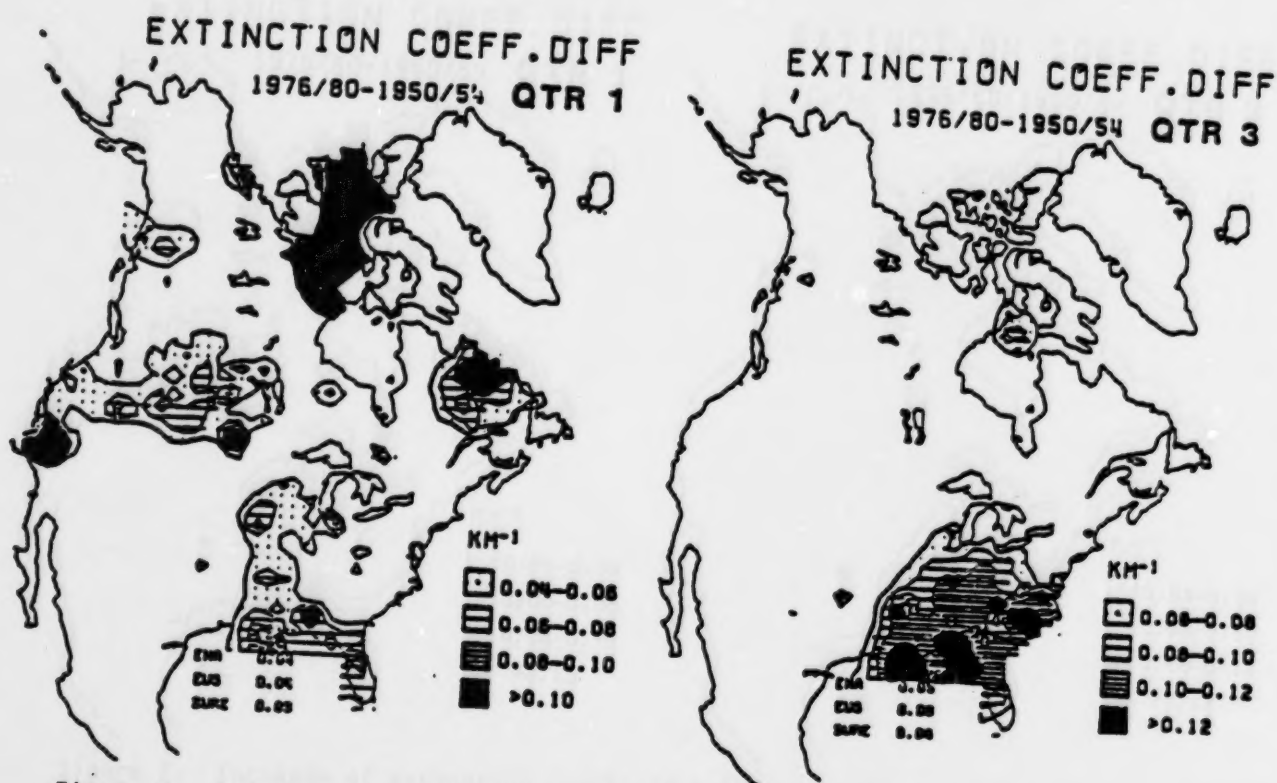


Figure 7. Increase of extinction coefficient from 1950-54 to 1976-80.
a) January, February, and March. b) July, August, and September.

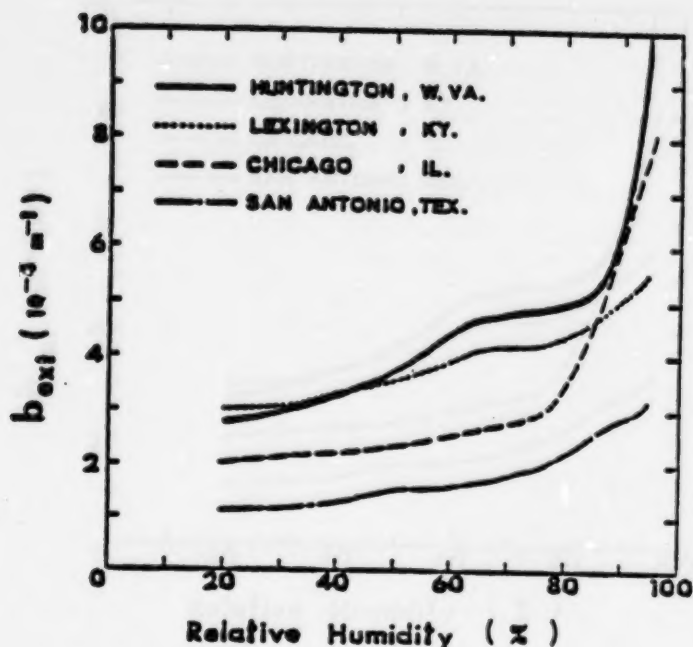


Figure 8. Humidity classified extinction coefficients for Huntington, WV, Lexington, KY, Chicago, IL, and San Antonio, TX (Husar et al., 1979).

The role of temperature as a factor influencing the haziness is difficult to perceive because of the many routes by which temperature dependence may be exerted. Even coarse examination of the existing historical data base reveals that major qualitative changes have occurred in the temperature dependence of haziness (Figure 9).

Wind speed influences the dispersion of particles, and if haziness is due to local sources (Washington, D.C., winter 1945-50 and see Figure 8), low wind speeds cause higher haze levels. For regional haziness (Washington D.C., winter 1970-75) the extinction coefficient is only weakly dependent on wind speed. In arid areas, e.g. Albuquerque, New Mexico, the extinction coefficient is the highest at high wind speeds because of the resuspension of dust (Figure 11).

These examples are intended to illuminate the vast reservoir of haze-related information contained in the existing meteorological data base once the data have been refined and standardized.

In an aerosol-free atmosphere, about one third of the solar energy passing through at a zenith angle of 60° (about 2 air masses) is depleted by Rayleigh scattering, water vapor and ozone absorption even in the absence of scattering and absorbing particles. On the order of ten percent of the lost direct

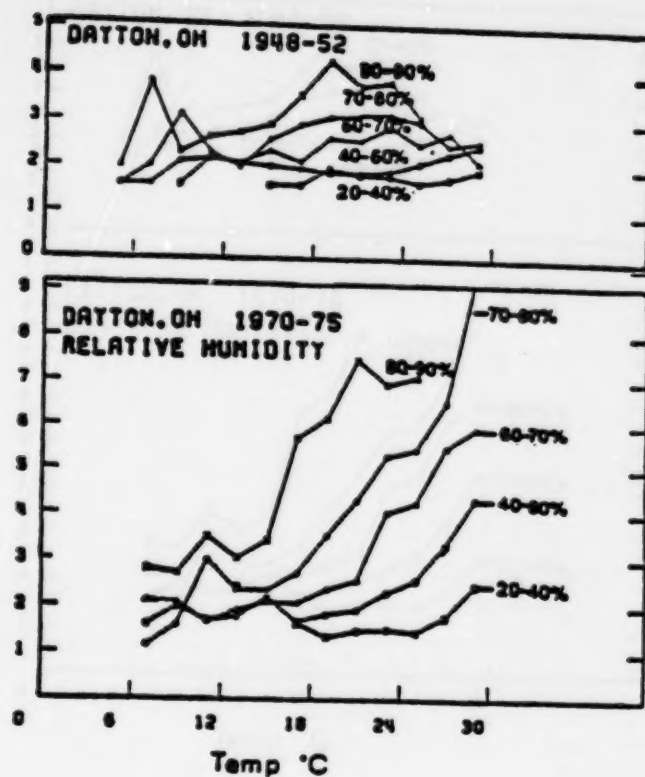


Figure 9. In Dayton, OH, 1948-52, the extinction coefficient was essentially temperature-independent while higher extinction coefficients were associated with higher RH. By 1970-75, the temperature-RH dependency of haziness has unfolded dramatically: high extinction coefficients are associated with high temperature and high relative humidity (Husar et al., 1979).

radiation is recovered as blue skylight while another 10% is backscattered to space to give the earth's atmospheric shell its bluish color as viewed from space. The remaining depleted energy heats the stratospheric ozone layer and the near surface layer containing the water vapor.

The scattering, ozone and water vapor absorption processes act on different parts of the solar spectrum as shown schematically in Figure 12. Furthermore, the atmospheric strata containing the ozone, air and water vapor are also separated.

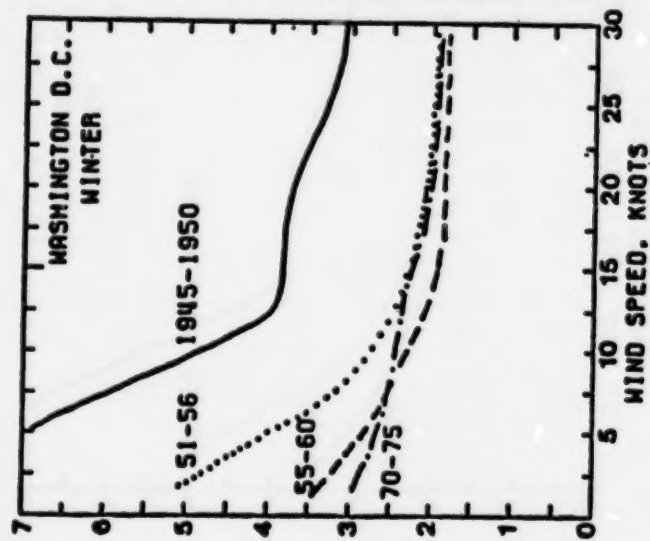


Figure 10. Wind speed dependence of extinction coefficient at Washington, D.C., winter 1945-50, 1951-56, 1955-60, and 1970-75.

Source: Husar et al. (1979).

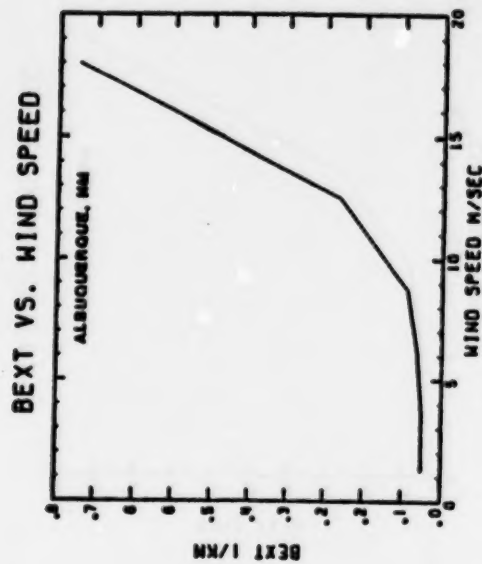


Figure 11. Wind speed dependence of extinction coefficient at Albuquerque, NM.

The incoming solar photons first encounter the ozone layer at 10-40 km above sea level. High energy photons in the UV ($<0.36 \mu\text{m}$) are absorbed in the strongly absorbing bands (Figure 13a) which, however, saturate quickly with increasing ozone depth. The absorption in the visible band, on the other hand, continues to increase almost linearly with ozone depth.

According to Figure 13, at air mass 2 and 3, respectively, 3 and 4% of the direct solar radiation is absorbed by ozone.

The second chance for the photon-atmosphere encounter occurs when the solar photons pass through the bulk of the atmosphere. Molecular scattering changes the photon direction mostly at the blue end of the spectrum since the scattering is λ^{-4} . Rayleigh scattering in solar radiation transmission is confined to $\lambda < 0.8 \mu\text{m}$.

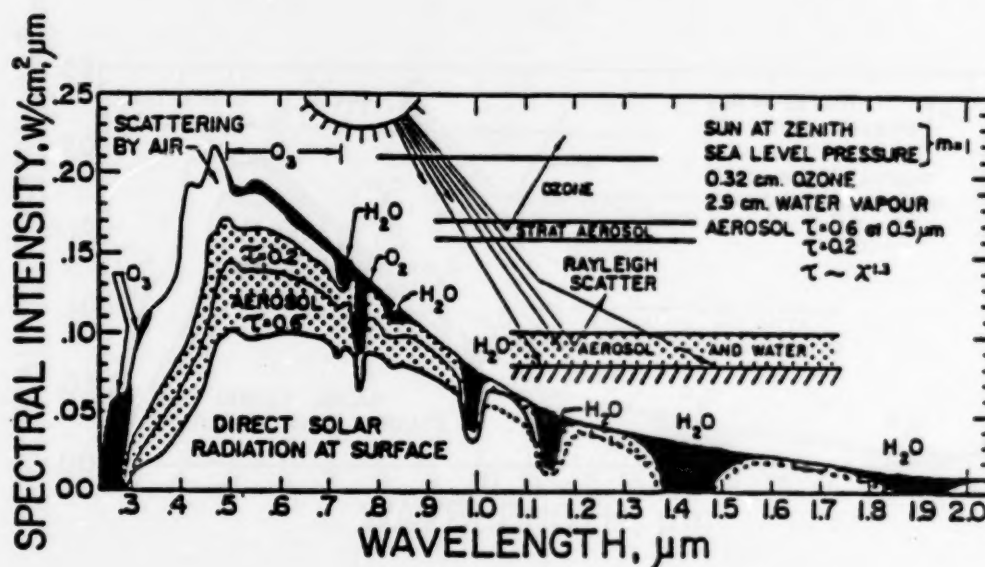


Figure 12. Illustration of solar radiation spectrum outside the atmosphere, and depleted by ozone, water vapor, Rayleigh scattering and aerosol. Parts adapted from Breslau and Dave (1973).

A third kind of encounter occurs when the solar photons reach the lowest layers of the atmosphere where most of the water vapor is contained. Water is a major "source" of atmospheric heating via its selective absorption of solar photons in the near IR bands (Figure 13). As shown in Figure 13b, for water paths >2 cm of precipitable water the increase in amount of solar radiation absorbed is markedly slowed due to the saturation of the main absorption bands.

Typical winter values of precipitable water content of the atmosphere are 0.5 - 1.0 cm in a vertical column over the eastern U.S.; in the summer it ranges between 2 and 4 cm. Estimates of the monthly precipitable water content and its standard deviation from one year to another at St. Cloud, MN is shown in Figure 13c (Reitan, 1960).

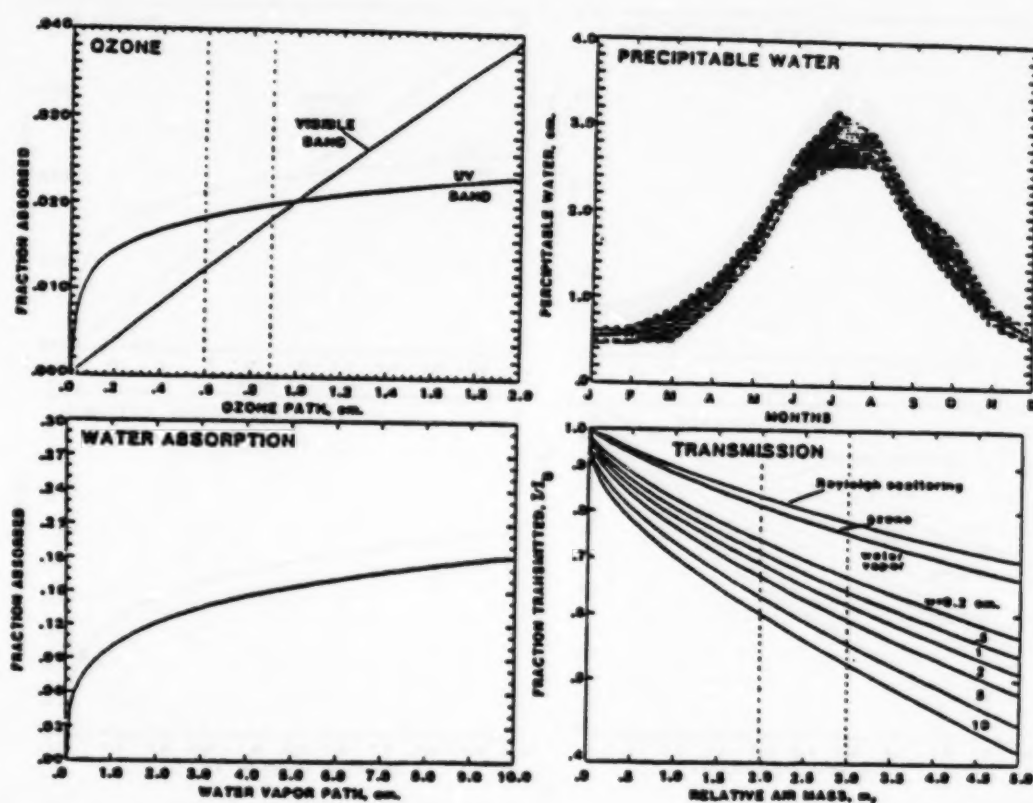


Figure 13. a) Ozone absorption. b) Water vapor absorption. c) Monthly precipitable water content and year-to-year standard deviations (shaded area) at St. Cloud, MN. d) Depletion of haze-free moist atmosphere at different values of precipitable water.

The abundance of aerosols in industrialized and even remote areas of the globe necessitates their careful consideration. The depletion by aerosols of the direct solar beam is estimated by the "difference," i.e. by comparing the values of extraterrestrial radiation appropriately depleted by O_3 and water absorption and molecular scattering to the observed ground level intensities. The excess depletion, beyond these known constituents, is attributed to aerosol scattering and absorption. This process is depicted in Figure 14.

Ever since Linke (e.g. Linke and Boda, 1922) and Ångström (1929), atmospheric radiation physicists have debated the question of how to account for the aerosols in calculating atmospheric transmission. The magnitude of the loss of irradiance (i.e. global radiation or total solar energy reaching the ground) and the internal consistency of the data set can be illustrated by an

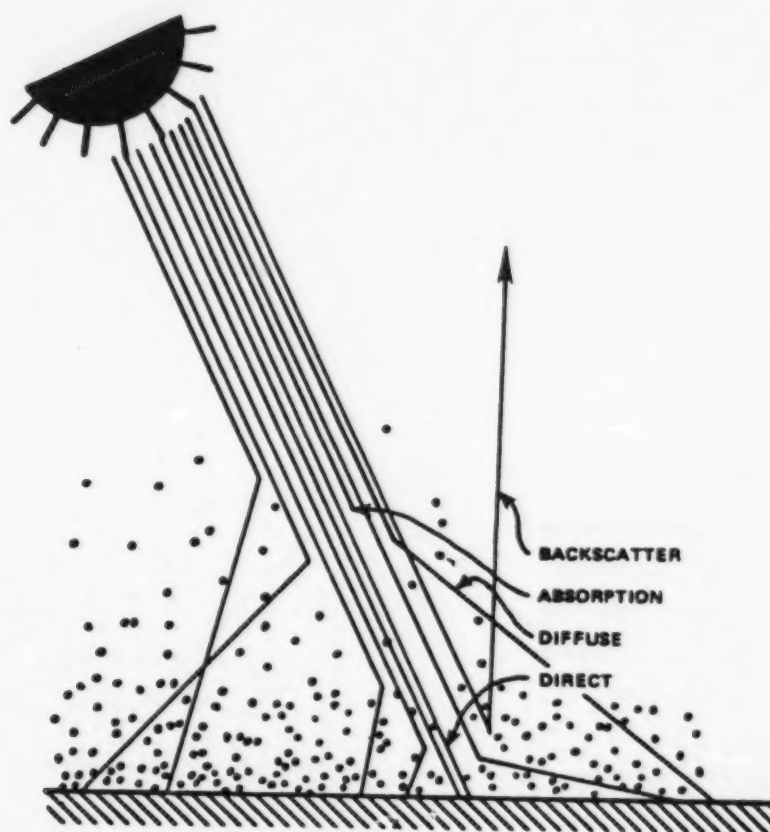


Figure 14. Extinction of direct solar radiation by aerosols is depicted.

example calculation. While what follows is primarily an empirical example, it can be justified by theoretical calculations of the optical effects via use of the aerosol size distribution and Mie computations. An average sulfate aerosol mass concentration of 10 mg/m^3 multiplied by a measured empirical scattering efficiency of $10 \text{ m}^2/\text{g}$ (Waggoner et al., 1982) yields an expected scattering component of extinction of 10^{-4} M^{-1} . This is in agreement with direct measurements and with visibility observations as well. A mixing depth of $\sim 2 \text{ km}$ for a layer with this scattering coefficient yields a scattering optical depth of 0.2. Since about 20% of the scattering results in a direct loss to space, this optical depth results in about a 4% loss of irradiance at the ground. Because the light absorption coefficient is often about 10% of the scattering coefficient, this causes an additional loss of 2% of irradiance, for a total loss of about 6%. This amount, deduced from typical microphysical measurements, is in agreement with direct measurements of solar irradiance at the ground (per Ball and Robinson, 1982). The absorption component of about 10% scattering is largely due to carbonaceous aerosols. Given that extinction/ fine mass concentration ratios for such absorbing aerosols typically range from 7 to $12 \text{ m}^2/\text{g}$ (Environmental Protection Agency, 1982), the elemental urban concentration can be estimated. Comparison to direct measurements of carbon are also consistent.

Regardless of the temperature of the earth as a whole, absorption of solar radiation by particles results in heating of the atmosphere and reduction in the energy reaching the earth's surface. Furthermore, with the exception of the stratospheric aerosol, any given type of particle (e.g. sulfate pollutant haze, windblown dust or sea salt) occurs systematically over small areas of the earth. Thus the direct effects of aerosols on radiative processes must be localized to these same regions. The horizontal spatial scales of relevance are variable, but generally fall into the range of a few hundred to a few thousand kilometers. It is also clear that the simple "warming versus cooling" analogy to CO_2 , which is very nearly uniformly distributed in the atmosphere, is too simple, both because of this geographical dependence and because of multiple but separate effects of particles. Cooling of the ground as a result of the loss of solar radiation (either by backscatter to space, or by absorption) is one effect, while heating of the atmosphere by absorption is another. A change in regional scale albedo is yet another effect.

Table 1 outlines some of the aerosol properties as observed at eleven urban sites, five rural, and three remote tropospheric (Charlson and Robinson, 1984; Waggoner et al., 1981). The eastern U.S. is almost continuously covered by a sulfate aerosol layer with concentrations averaging around $10 \mu\text{g}/\text{m}^3$. A several-state area shows maxima in annual averages of sulfate concentration approaching $15 - 18 \mu\text{g}/\text{m}^3$. Given the range of values for the ratio of $\sigma_{\text{sp}}/[\text{SO}_4^{2-}]$, annual average scattering extinction coefficients of 10^{-4} , $^{-1}$ or larger might be expected. Ball and Robinson (1982) recently noted that haze (almost entirely anthropogenic) in "central regions of the United States reduces the annual solar irradiation of the surface by $\sim 7.5\%$."

On a regional scale, the extinction of light is generally dominated by particle scattering. Most aerosol particles, including anthropogenic material, are introduced into the atmospheric boundary layer. Under free convection, the boundary layer becomes well mixed, its potential temperature, water vapor mixing ratio and particle content tending to assume a slowly changing distribution with height. Examination of atmospheric soundings downwind of source regions, whether over ocean or land, tend to show that the dust layer can be identified as a well mixed stratum, typically with a base at 1-2 km and a top close to the level of the top of the source mixing layer, which may reach altitudes of 4-6 km. This elevated mixed layer is typical of boundary layer air which has been advected from its source (Carlson, 1984 meeting).

If there are no clouds between the observer and the sun, the intensity of direct solar radiation for a given solar elevation depends on the variable amount of dust, haze, and water vapor in the atmosphere. The extinction produced by these constituents is called "atmospheric turbidity." During hazy episodes, turbidity coefficients as high as 0.6 to 1.0 have been reported, translating into a 75 to 90 percent removal of solar radiation from the direct beam, a 7.5 to 9 percent loss to space, and a 7.5 to 9 percent loss at atmospheric heating.

Data from a 29-station turbidity network showed that there are strong spatial and temporal variations of turbidity across the United States. At all 29 sites, the highest turbidity occurs in the summertime and the lowest occurs in the winter, which is consistent with the haziness pattern obtained from visibility observations. The turbidity of the atmosphere in the United States has a strong spatial dependence. In the Southwest, with a mean annual turbidity coefficient of about 0.06, the incoming direct solar radiation is attenuated by only 13 percent, compared with Midwest values of about 20 percent.

TABLE 1. AEROSOL PROPERTIES AT VARIOUS URBAN, RURAL, AND REMOTE SITES

A.	Site Urban	Scattering Extinction $\pm$ standard deviation $\sigma_{spg} \pm s_g$ (m^{-1})	Back- Scatter Ratio R(90%)	Albedo for Single Scatter ω_g	Number of Determinations N
1.	Phoenix, AZ 11-17-78/11-23-78	3.0E-4 1.9E-4 1.2E-4	--	0.68 0.63 0.59	20
2.	Denver, CO (Henderson) 11-10-73/11-15-73	9.7E-5 3.4E-5 1.2E-5	0.15 0.12 0.11	0.69 0.50 0.37	13*
3.	Denver, CO (Trout Farm) 11-15-73/11-21-73	1.4E-4 5.8E-5 2.3E-5	0.13 0.11 0.09	0.67 0.54 0.43	23
4.	Denver, CO 11-20-78/12-20-78	1.8E-4 6.2E-5 2.2E-5	--	0.73 0.61 0.49	103
5.	Seattle, WA (Industrial) 3-18-77/4-10-77	9.7E-5 3.6E-5 1.4E-5	--	0.76 0.65 0.55	43
6.	Seattle, WA (Residential) 5-24-77/6-11-77	5.8E-5 3.5E-5 2.1E-5	--	0.87 0.78 0.70	69
7.	Portland, OR 1-16-77/3-2-77	2.1E-4 7.4E-5 2.7E-5	--	0.67 0.55 0.45	159
8.	St. Louis, MO (St. Louis Univ.) 9-28-73/10-4-73	1.1E-4 6.5E-5 3.7E-5	0.11 0.10 0.07	0.70 0.59 0.49	50
9.	St. Louis, MO (Wash. Univ.) 8-22-73/8-30-73	1.9E-4 1.2E-4 7.5E-5	0.10 0.08 0.07	0.86 0.76 0.67	50
10.	New York, NY 12-14-67/12-22-67	7.3E-4 4.3E-4 1.3E-4	--	0.72 0.62 0.52	9*
11.	Riverside, CA 11-05-75/11-07-75	1.1E-3 8.9E-4 7.9E-4	--	0.94 0.93 0.91	6*

TABLE 1. (continued)

B.	Site Rural	Scattering Extinction ± standard deviation $\sigma_{\text{spg}} \pm s_g$ (m^{-1})	Back- Scatter Ratio R(90%)	Albedo for Single Scatter $\tilde{\omega}_g$	Number of Determinations N
1.	Milford, MI 10-1-75/10-14-75	7.1E-5 3.4E-5 1.6E-5	0.17 0.14 0.11	0.80 0.72 0.65	8*
2.	Hall Mt., AR 11-6-75/12-4-75	6.2E-5 4.2E-5 2.5E-5	0.17 0.11 0.07	0.95 0.86 0.80	62
3.	Puget Island, WA 3-5-75/3-17-75	3.6E-5 1.5E-5 6.1E-6	--	0.89 0.79 0.71	23
4.	Tyson, MO 9-5-73/9-26-73	1.4E-4 6.5E-5 2.9E-5	0.14 0.09 0.07	0.94 0.78 0.64	96
5.	Tyson, MO 8-13-75/9-26-75	1.2E-4 4.9E-5 2.1E-5	0.14 0.10 0.06	0.90 0.79 0.70	52
C. Remote, Lower Troposphere					
1.	Above Flagstaff, AZ 10-30-74/11-6-74	1.9E-5 1.7E-5 1.4E-5	--	0.97 0.94 0.91	4*
2.	Mesa Verde, CO 9-13-78/9-20-78	3.3E-5 1.8E-5 1.0E-5	--	0.94 0.90 0.85	4*
3.	Abastumani Obs, USSR 7-4-79/7-26-79	9.9E-5 5.3E-5 2.9E-5	--	0.92 0.90 0.87	40

*No s_g given due to small number of points. Limits are simply extremal values.

Source: Weiss, 1980

The highest turbidity coefficients were observed in the Eastern United States (Figure 15) where winter values of 0.1 and summer values of 0.2 were typical, meaning that about 20 to 35 percent of the direct solar beam is diverted to sky light, backscattered to space, or absorbed.

In the 1970's, the U.S. turbidity network was expanded under the auspices of the World Meteorological Organization (WMO). In the northern hemisphere there were about 100 operating stations. As with every network, the quality and quantity of data from individual stations are highly variable. The U.S. turbidity network has been deteriorating since 1976, and it is not certain whether it will survive the decade of the 1980's.

Aerosol layers are transported via the boundary or mixing layer. The aerosol properties can be identified downstream in a differing configuration after inversion at the boundary line. Virtually all atmospheric aerosols, natural or manmade, reside in a mixing layer, which tends to possess relatively uniform and identifiable characteristics. These characteristics can be traced for large distances - often more than 1000 km.

As one example, trajectories computed using the ARL-ATAD model for the month of July 1982 were used to determine the direction from which the air traveled to Whiteface Mountain, New York. Measurements were assigned the computed trajectories closest to the time the measurements were made, with only the first 48 hours of the computed trajectories used to determine the sector of approach.

There is considerably more oxidized sulfur and nitrogen observed in air originating in the Southwest than in air originating from the Northwest. Also, concentrations of both sulfate and SO_2 are greater in the southwest sectors than the northwest sectors, but the difference is more pronounced for sulfate than for SO_2 . There is 10 times as much sulfate, on the average, in air coming from the sector between 210 and 240 degrees than that between 330 and 360 degrees, for example, but only four times as much SO_2 comparing these two sectors. There are also higher nitric acid and NO_x concentrations in air coming from the southwest sectors than the northwest sectors, but the difference is more pronounced for nitric acid than for NO_x . Much higher aerosol ammonia concentrations are found in the southwest sector than in the northwest sector, reflecting the larger sources of ammonia in the agricultural regions of the midwest and the higher concentrations of sulfate in the aerosols coming from this region. There also is considerably more aerosol acidity in air

coming from the southwest sectors. This increased aerosol acidity is due to the larger amounts of sulfate observed in these sectors, some of which is present as sulfuric acid.

Recent studies of the gas phase, aerosol and cloud mechanisms of acid generation in the troposphere suggest a variety of contributing reaction pathways in the transformation of air pollutants.

For photochemical systems, there appears to be a marked dependence of the rates on the contamination of the air mass. The maximum theoretical rates of SO_2 oxidation occur with relative clean air masses. These rates are far less than the instantaneous rates estimated for the cloud water reactions, but during a 24-hour cloud-free period they can result in substantial conversion of the initial SO_2 to acid.

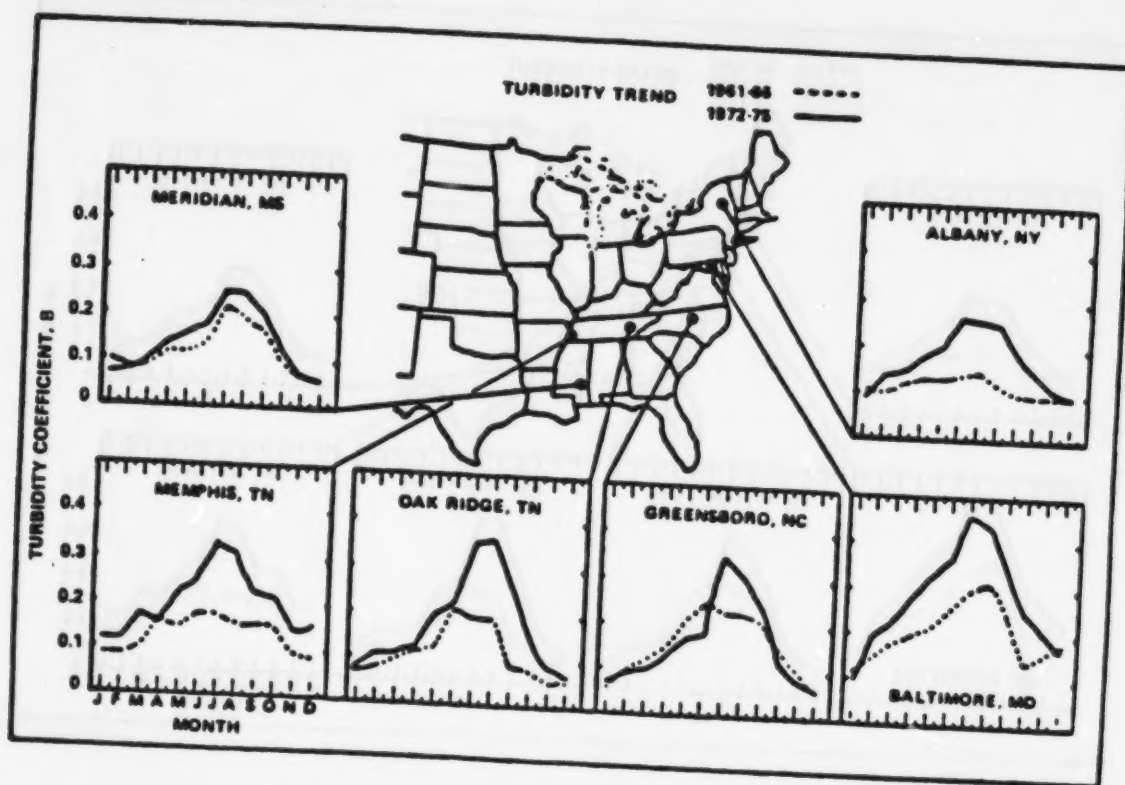


Figure 15. Seasonal turbidity patterns for 1961-1966 and 1972-1975 are shown for selected regions in the Eastern United States.

Source: Flowers et al. (1969), and WMO (1974 through 1977).

Clearly the homogeneous chemistry of the troposphere can lead to significant average rates of acid generation for the clear summertime months over times of a few days and transport distances of a thousand or more kilometers. The fractional contribution which such homogeneous processes make to the total acid which is deposited in sensitive areas is uncertain. If account is taken of the fractions of the time that the troposphere is relatively cloud-free and cloud-rich, then the qualitative conclusion which one can make is that the contributions from both gas-phase and cloud reactions are significant.

The global cloud cover plays a vital role in the earth's radiative budget in reflecting energy back to space, in absorbing both solar and longwave (terrestrial) radiation, and in emitting its energy downward and outward into space. Changes in cloud cover, therefore, alter the global heat balance. Cloud-and precipitation-forming processes may be divided into two broad classes: (1) macrophysical processes, which affect the rise and descent of air currents and the amount of water vapor available for condensation; and (2) microphysical processes, which affect the nature of cloud particles formed during condensation. The role of atmospheric aerosols, primarily those that are strongly hygroscopic, is thought to influence primarily the microphysics of cloud formation.

On a global or even regional scale, the very small amounts of moisture that man adds by land practices or combustion of fossil fuels are negligible in comparison with global evaporation. On a regional scale, only one form of increasing cloudiness suggests itself: the formation of aircraft contrails (Machta and Telegadas, 1974). Aircraft contrail formation results mainly from the injection of water vapor rather than of aerosols.

Oxidation of SO_2 to $\text{SO}_4^=$ inside droplets increases the amount of particulate matter in the atmosphere. Moreover, clouds redistribute existing particulate matter. This may modify the temperature profile and hence onset, duration, and/or rate of precipitation. Also, we now know transport of materials inside clouds can bring purer, unmixed cloud air from cloud base to the top of thunderclouds. On the other hand, material entrained at cloudtop can mix with cloud air to form negatively buoyant parcels which can descend almost to cloud base. Dissipation of clouds during this transport process (and initiated by entrainment of dry air) results in a release of gases and aerosols into non cloudy air. All these processes have very important impacts on redistribution of "contaminants," not only in the troposphere, but up to the tropopause and

beyond in some cases. These materials are not only the original particles which entered at cloud base, but clearly any products of liquid-phase chemistry due to incorporation of gases during entrainment.

Note that in the course of the transport of cloud base and cloud top materials up and down, there may be modification of ice-phase processes which can reduce ice particle multiplication with attendant effects upon electrification. These phenomena are of interest in precipitation and cloud formation and constitute an area requiring further study.

The important question of the influence of aerosols (including light absorbers) on the albedo of clouds must still be left as an open question. We cannot say whether (1) the primary effect of increased amounts of aerosol is a higher concentration of cloud droplets leading to increased albedo or (2) whether the presence of light-absorbing matter (carbon) causes a decrease in albedo. In any case there is good reason to suspect that both local and global climate could be very sensitive to such influences.

Summarizing the results of various observing techniques, we see that the existing data show a high degree of internal consistency. Several key conclusions emerge:

1. Fine particle aerosols cause the extinction of visible radiation.
2. The dominant species in fine particle aerosols is sulfate and it accounts for a significant fraction of the extinction due to scattering, perhaps half on the average in the eastern U.S.
3. The dominant light absorber in these aerosols is elemental, graphitic carbon which originates as soot. Light absorption is large in urban areas and still exists at measurable levels in rural areas of the eastern U.S.
4. Chemical data ($\text{SO}_4^{=}$ and C^0) are consistent with optical data when linked with the Mie formalism and aerosol particle size distribution.
5. The local scattering and absorption extinction data are consistent with extinction determined from either visual range or turbidity.
6. Most of the eastern U.S. is influenced by these aerosols; the effects are not just urban or local. In cities, higher values of extinction and especially of absorption indicate larger effects may be expected there.
7. The pattern is also seasonal, e.g., in New England there is improvement in winter and decrease in summer, whereas in the Smokies the winter level remains steady and in the summer it becomes much worse.

8. In light haze situations the downward atmospheric radiation flux increases slightly because of the rise in temperature of the particle-containing layer, in spite of the decrease in water content which follows the computed decrease of evaporation.
9. Aerosols do go long distances (dispersed), but are not uniformly distributed over the globe. A typical lifetime is 3 - 5 days and a typical transport distance is 1000 km.
10. Transport models and models of entrainment and precipitation that link clear air aerosol concentrations and cloud physics have been developed.
11. Entrainment and transport may redistribute materials both up and down.

Direct effects such as these, along with influences on cloud nucleation, result in indirect effects such as changes in convective activity, changes in cloud distribution in time or space, and perhaps shifts in synoptic patterns. This picture of increasingly complex effects stemming from simpler ones leads to the general notion that there is a hierarchy of effects, all of which are based on a few primary physical interactions.

IV. Impact on Climate.

The direct effects described above (Section III) have indirect influence on a variety of phenomena, which for purposes of this paper are grouped under a discussion of climate. Observations about climate require taking a systematic view of the interactions between pollutant patterns and parameters such as temperature fluctuations, precipitation, dew point, humidity, and cloud formation.

It has long been recognized that the amount of solar radiation reaching the surface of the earth is an important climatological variable. Abbott (1911) and others demonstrated the near constancy of the amount of solar radiation reaching the outer limits of the atmosphere--as is implied by the use of the term "solar constant." Indeed, study of solar variability as a cause of climate change is still an active field of research. At the present time, however, attention must be drawn to the fact that the variations in radiation reaching the surface of the earth are to a much larger extent due to fluctuations in the transmission power of the atmosphere than to the variations of the solar constant itself.

McCormik and Ludwig (1967) and Bryson (1968) presented arguments to show that a buildup of atmospheric aerosols could have the effect of increasing albedo, thereby cooling the earth. Charlson and Pilat (1969) suggested that since particles can both absorb and scatter light, particles could either heat or cool the earth, depending on the relative amounts of absorption and scattering. Atwater (1970) discussed factors governing the sign of the change in the planetary albedo caused by the presence of aerosols, showing that either heating or cooling could result. Of particular importance is the albedo of the surface under an aerosol layer. Mitchell (1971) also noted the roles of both scattering and absorption, with similar conclusions depending on albedo, but with some adjustment for evaporation of water at the surface. Ensor et al. (1971) developed a model that allowed the imaginary refraction index to be varied, concluding that large ambiguities exist due to a lack of data on the absorption and scattering properties of aerosols. Chylek and Coakley (1975) utilized a two-stream approximation and dependence on the angle of scattering to produce estimates of aerosol effects for optically thin atmospheres. As a result, they predicted that cooling would occur in arctic regions.

Reck (1974, 1975, 1977) explored a variety of the parameters that are imbedded in models of the influence of aerosols, suggesting that aerosols cool the atmosphere, reduce convection and increase planetary albedo. Hobbs et al. (1974) suggested that pollutants affecting clouds are the most likely to produce modifications in weather and climate.

Robinson (1977) discussed in general the effects of the particulate matter associated with energy production, focusing on surface temperature, planetary albedo and atmospheric heating. A major conclusion was that energy production, e.g., from burning sulfur-containing coal, resulted in a net energy loss from the earth's surface. That is, more solar energy is absorbed in the atmosphere and reflected into space than is released by the burning of coal.

The investigations listed above are in the main concerned with potential changes in the climate of the whole earth. There are many other aspects of climate and climate change, manifested over a wide range of time and space scales. For example, a substantial change might occur in the diurnal range of temperature in a semi-desert region. There need be no change in even the local daily average temperature, and certainly no change in any global statistic, but the local climate would have changed in a manner highly significant to life in the region.

Part of this section reports on work done by Husar to analyze the continental scale trends for climatic parameters that are potentially influenced by atmospheric haze and vice versa. The goal was to explore whether any significant regionalized climatic changes had occurred in the 1950-80 period.

This analysis drew upon a 30-year data base of 3-hourly surface meteorological observations for about 600 stations covering the U.S. and Canada. The data base utilizes computerized weather records supplied by the NOAA National Climatic Center and by the Atmospheric Environment Service Canada. (See Figures 16 and 17).

To summarize the findings, comparisons on data from 1950-54 and 1975-79 indicate a reduction in northeastern noon surface temperature of 1°C (no change in western U.S. and Canada) and an increase in midnight temperature, coinciding with increased aerosols in the atmosphere. Also, there have been changes in the dew point from 16.7°C to 18.0°C and an increase in relative humidity from 54.5°C to 60.5°C for the eastern U.S. During the same period, there has been an increase in cloud cover in the lower layer of the atmosphere (the same layer where haze occurs) of 30% to 35%. Fog frequency doubled from 7% to 14%, and total noontime cover increased about 5%, from 58% to 63%. Though there was no observable change in frequency of precipitation, there was some increase in the summertime pressure field, thus bringing in more moisture. In sum, there is now a consistent data base showing climatic changes in the northeastern U.S. These climatic changes correspond to a period of increased haze. The challenge to researchers is to justify scientifically the attribution of these climatic changes to the increase in the amount of haze in the region.

The following is a brief overview of the current research issues related to the climatic factors included in the above discussion.

The presence of aerosols in the mixing layer leads to a number of perturbations in the radiative energy balance. The aerosol layer over the eastern United States may cause, on a yearly average, a loss of about 7% of the solar energy which would otherwise reach the ground. About one-half is lost by backscatter to space. The other half is absorbed within the boundary layer with less heating of the ground and more heating of the air in the boundary layer. Heat loss through emission of long wavelength radiation by aerosols may or may not balance energy absorption in the visible wavelengths. If one process dominates the other, the aerosol may lead to either net cooling or a net heating of the boundary layer.

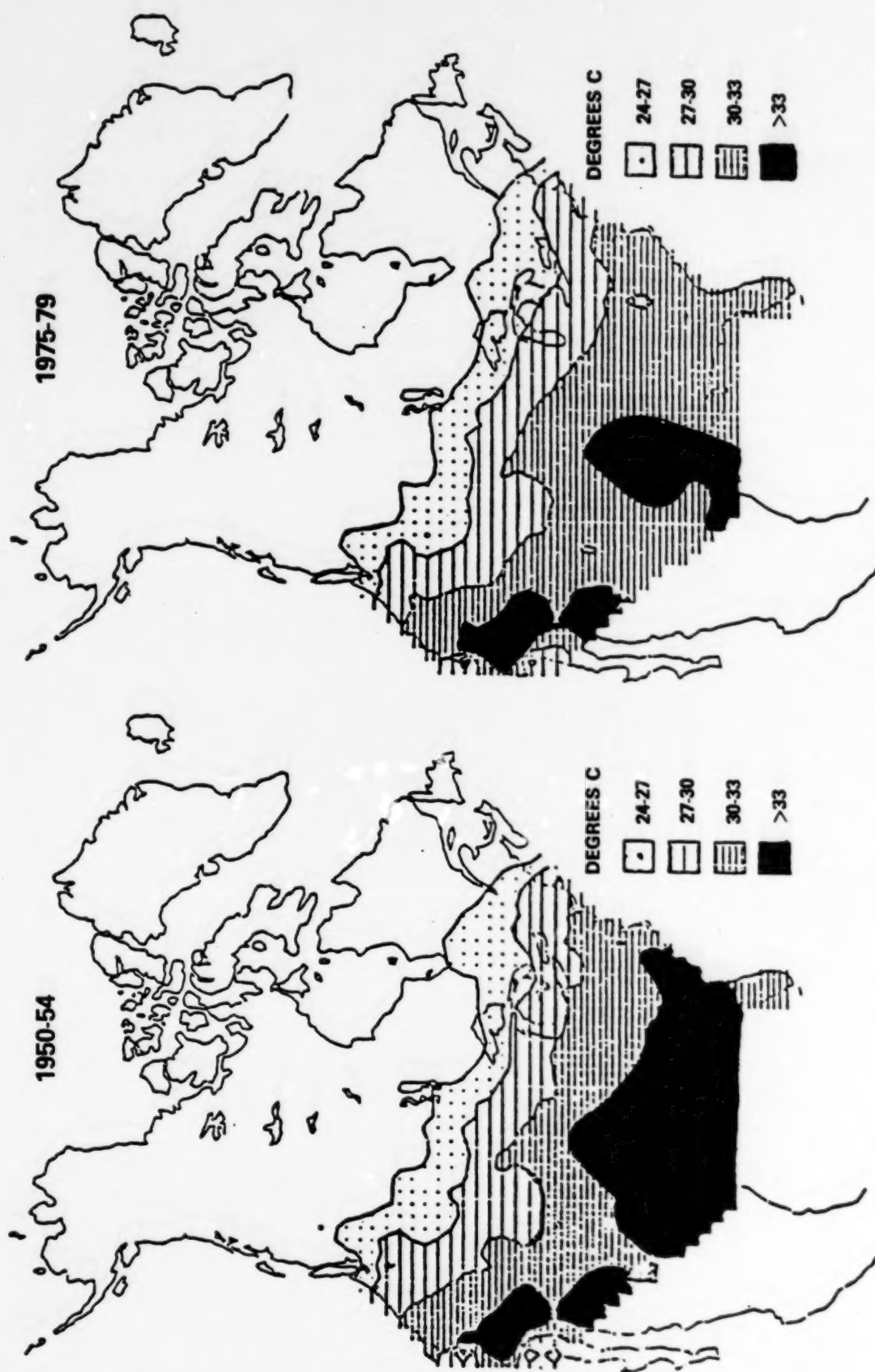


Figure 16. Noon temperature contour maps. Summer averages.
(3rd quarter)

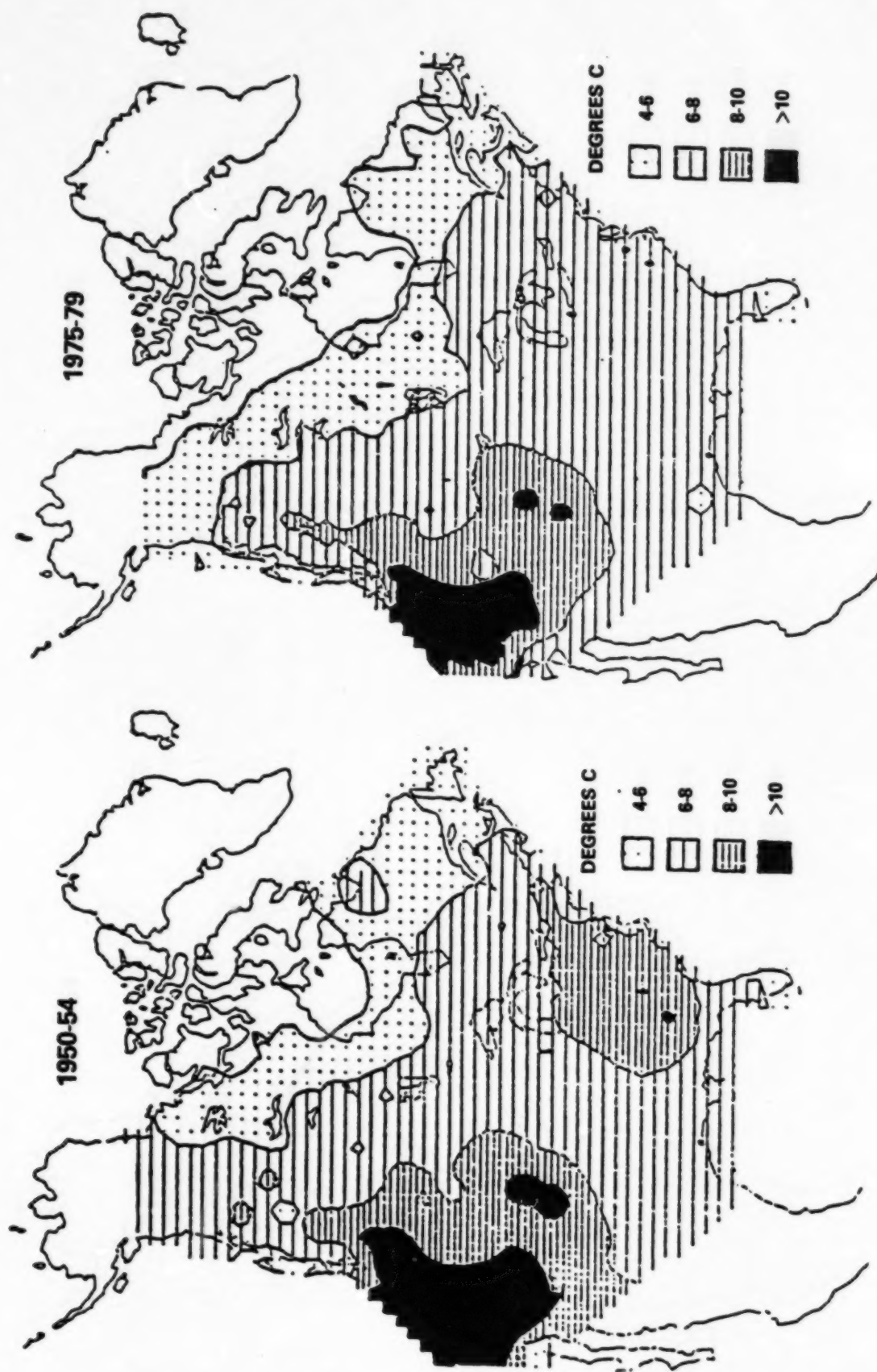


Figure 17. The average difference between noon and midnight temperatures. Summer season. (3rd quarter)

The most remarkable 30-year climatic change is noted for dew point (Figure 18). In the 1950's only a narrow belt of the Gulf coast recorded noon average dew point in excess of 20°C. By the 1970's this high dew point belt had expanded several hundred kilometers inland from the Gulf coast. For the entire eastern U.S. average this resulted in a dew point increase from 16.3 to 18 C. Here again it is worth emphasizing that neither the western U.S. or Canada exhibits such trends. It is thus possible that experimental errors are responsible for the trends.

The observed reduction of the noon temperature coupled with an increase of dew point resulted in a substantial increase in noon average relative humidity (Figure 19). In the 1950's virtually the entire U.S. had a noon average RH below 55%. By the 1970's the entire eastern U.S. exceeded this RH. Thus, on the average, the eastern U.S. noon RH has increased from 54.6 to 60.5%.

At present a plausible explanation is lacking for these anomalies. However, the significance of the remarkable increase in the water content of the surface atmosphere can not be overemphasized.

Aerosols have an influence on the optical properties and microphysics of clouds. The increased number of cloud condensation nuclei due to pollution aerosol can increase the number of cloud condensation nuclei resulting in an increased albedo. Carbon particles are present in rain, snow, and ice, and are therefore expected to be in cloud droplets. The presence of carbon should lead to a decrease in albedo.

Secular trends are presented here for two measures of cloud cover (Figures 20 and 21). The noon cloud cover reported by the U.S. weather stations refers to the percent of the sky covered by clouds within the lowest 1-2 km of the atmosphere. Canadian surface observations do not contain this parameter causing a sharp gradient at the U.S.-Canadian border (Figure 20). The noon low level cloud cover east of the Mississippi was 31% and increased to 35% by the 1970's. The increase was again most pronounced in the southeastern states.

The maps of the north American noontime total sky cover are depicted in Figure 21. The pattern shows an increase from 58 to 63% for the eastern U.S.

The estimation of the low level and total cloud cover is made subjectively by individual observers. It is thus conceivable that an increased haziness of the sky prompted observers to overestimate the sky cover in the 1970's. On the other hand, the increased cloud cover is consistent with the measured increase of atmospheric moisture over the southeastern U.S. If, in fact, the

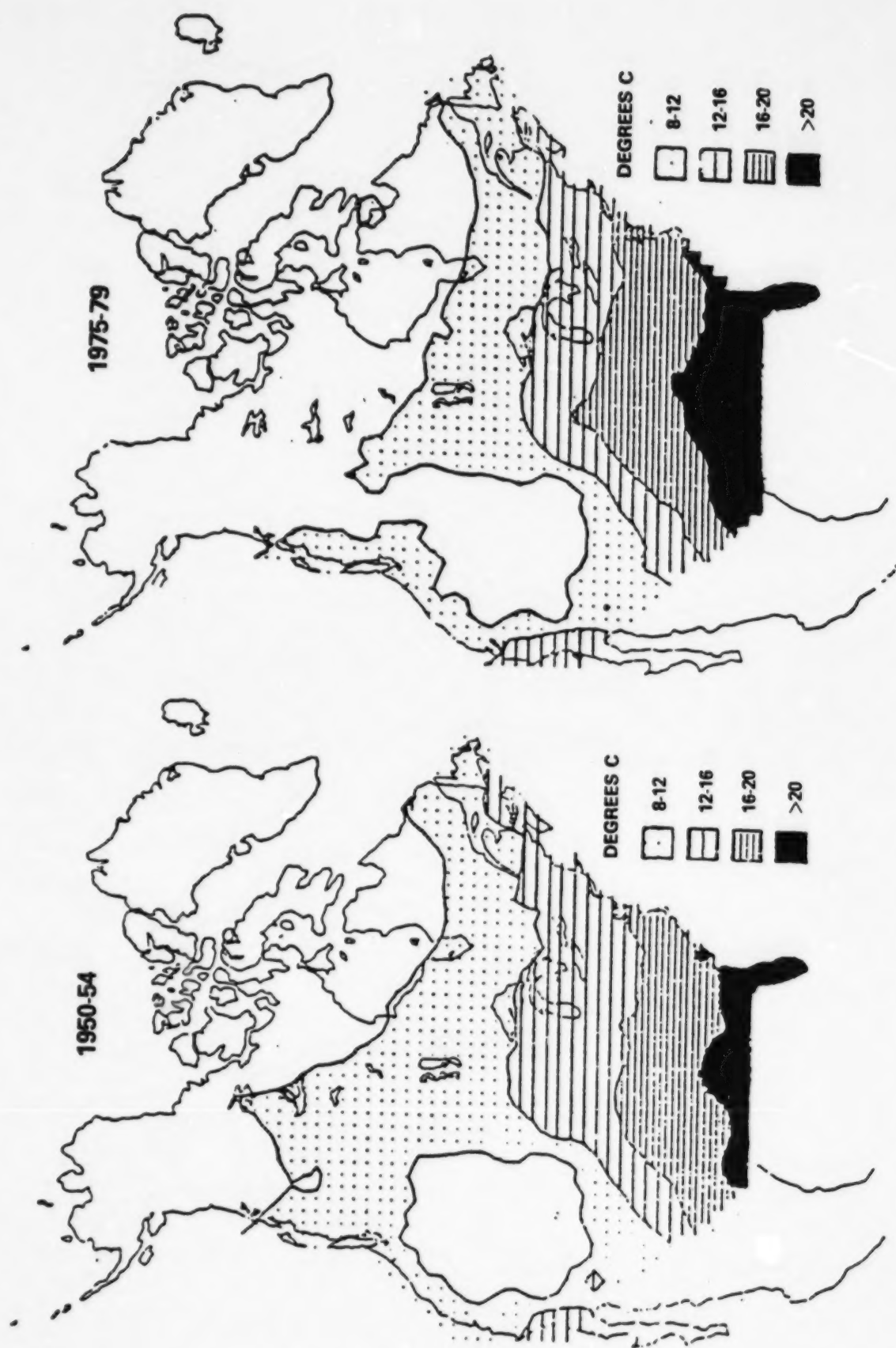


Figure 18. Noon average dew point in °C. Summer season.
(3rd quarter)

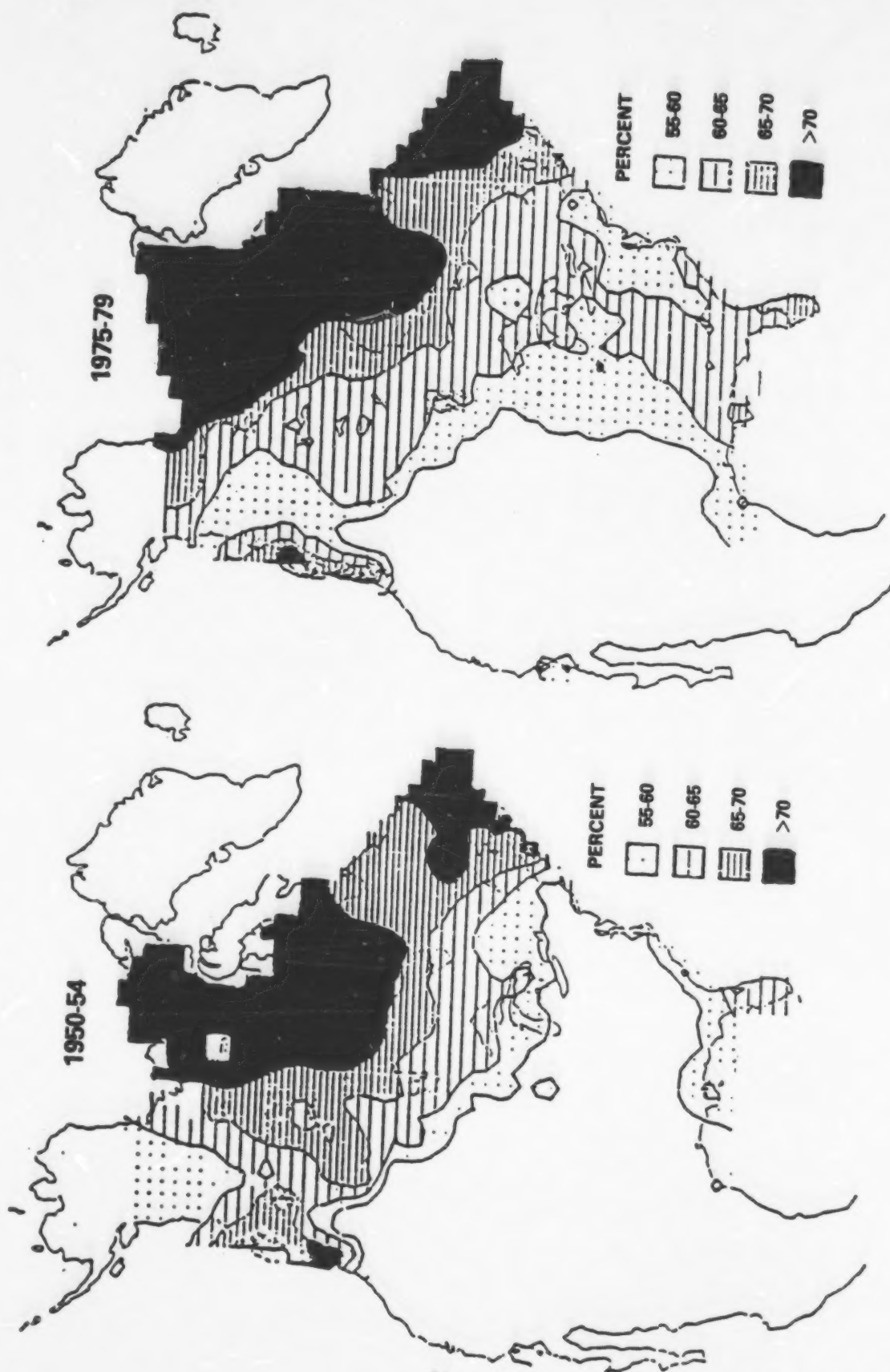


Figure 19. Average relative humidity. Summer season. (3rd quarter)

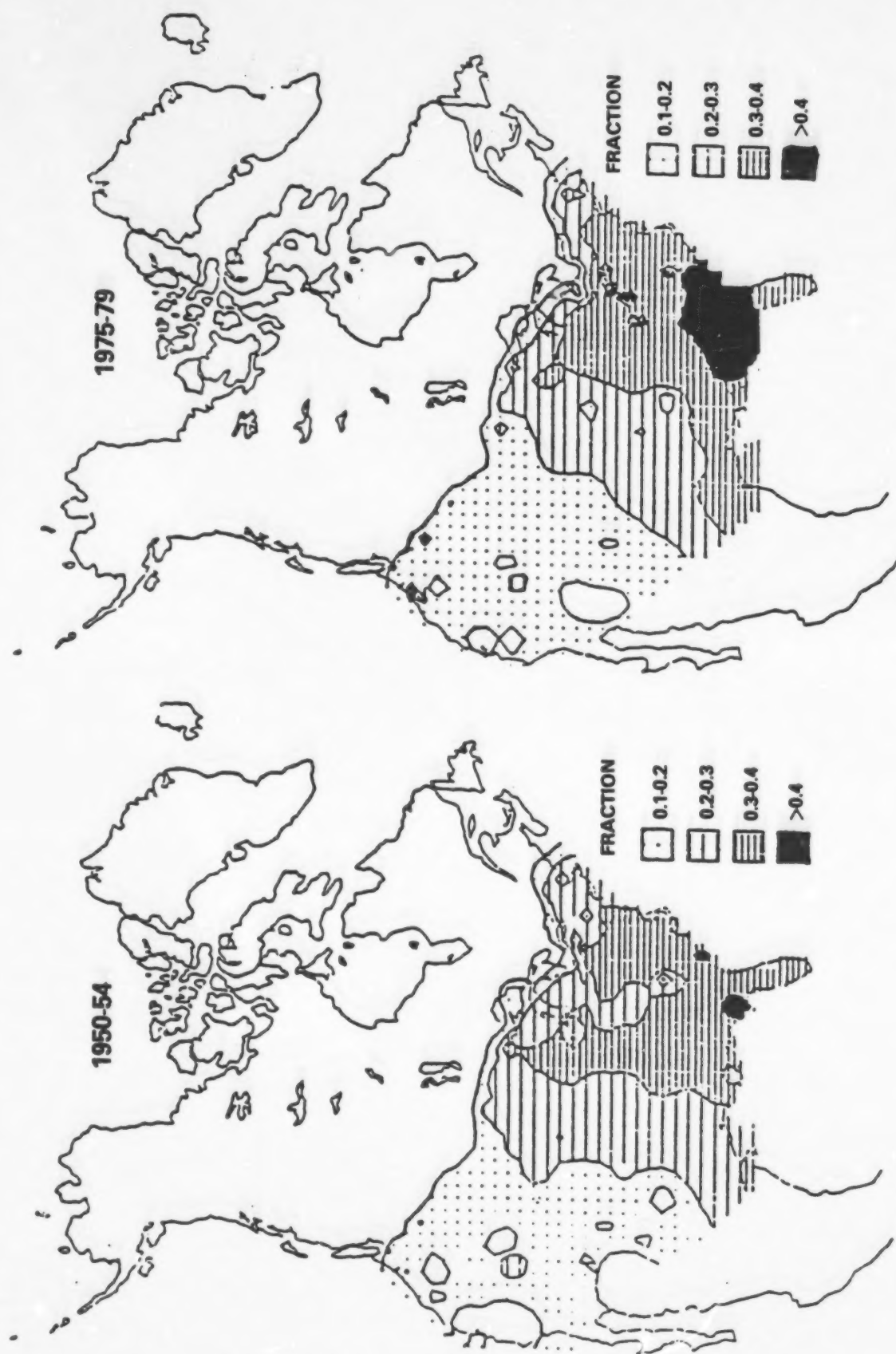


Figure 20. Noon low level cloud cover. Summer season.
(3rd quarter)

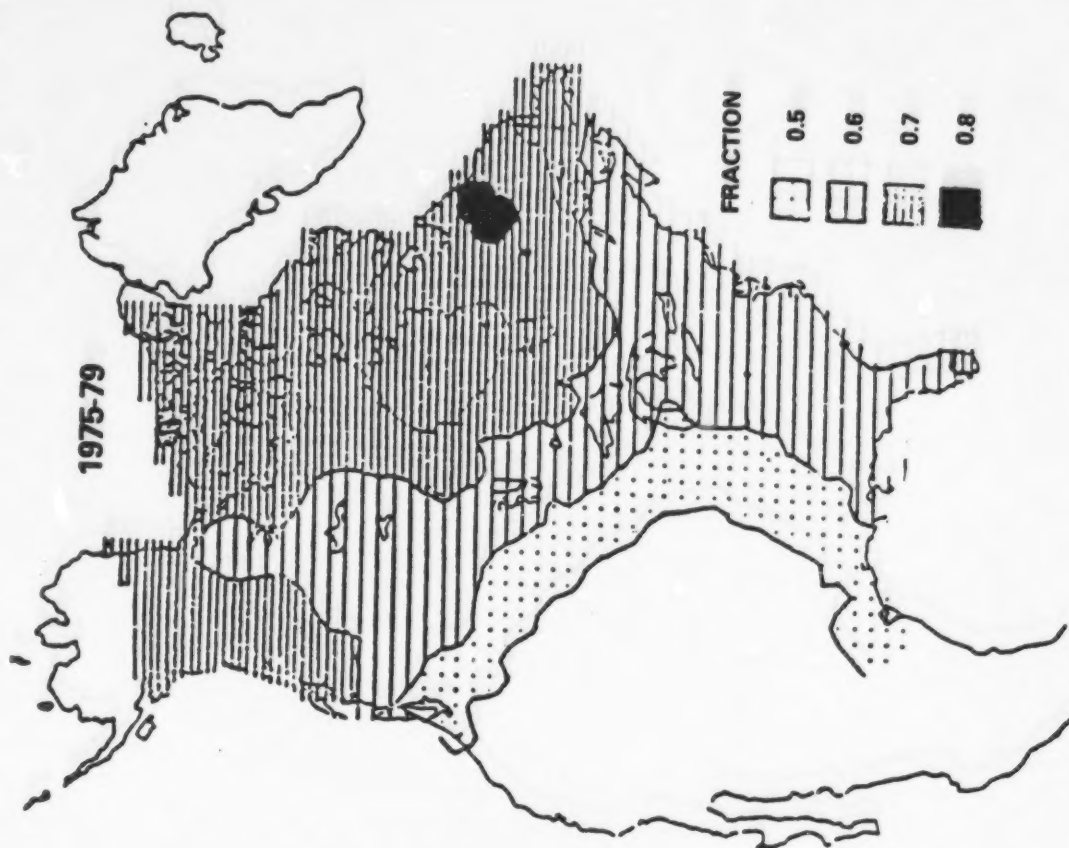
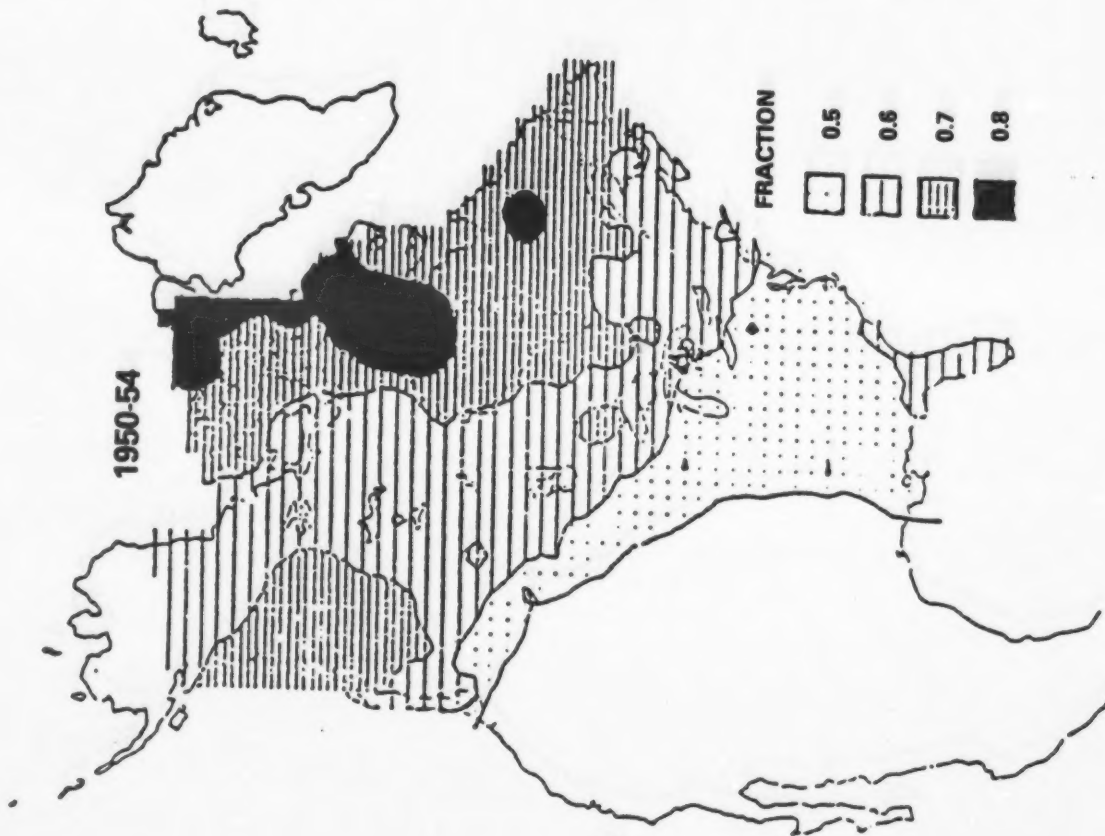


Figure 21. Noon total cloud cover. Summer season.
(3rd quarter)

eastern U.S. cloud cover has increased by 5% over the past 30 years, it could explain both the temperature reduction and demodulation, and the effect of aerosols could be nil.

The observation of fog is again a subjective judgement made by the observers. It generally refers to visual range less than 0.5-1 km. The North American map of reported fog frequencies shows that in the 1950's only the New England states, Quebec and Nova Scotia have experienced fog more than 10% of the time. By the 1970's virtually the entire eastern North America had experienced fog more than 10% of the time. The eastern U.S. average fog frequency had doubled from 7 to 14% of the 3-hourly observations (Figure 22).

The precipitation frequency is mapped in Figure 23. It is evident that the rainiest regions are in Canada (>20% of the time), while the least rainy is the arid southwest (<5% of the time). The eastern U.S. raininess does not show a significant secular trend.

A potential cause of secular climatic trends is a change in the regional scale circulation pattern. Currently we do not possess a convenient measure of the regional moisture and heat transport field. A possible surrogate for this purpose is the sea level pressure field depicted in Figure 24. The main features of the North American pressure field were similar in the 1950's and 1970's, although there was a slight increase of the eastern U.S. average from 1016.7 to 1017.3 mb. It is unclear whether this magnitude of pressure change might be responsible for the observed temperature, moisture and cloud cover trends.

The complex interactions of cloud- and precipitation-forming processes, however, obscure the specific role of manmade aerosols. Accordingly, climatic effects cannot at this time be quantitatively and causally related to pollutant emissions or to aerosol concentrations. We can say with confidence, however, that the aerosol has caused a decrease of irradiance at the ground of about 7%.

Unfortunately, there is little agreement about whether the net effect of increased airborne particle concentrations is the warming or cooling of the earth as a whole. Most models can predict either an increase or a decrease in the effective albedo of the earth under cloudless skies, depending on which combination of surface albedo, sun angle, particle size distribution, and particle complex refractive index is assumed. The effects of clouds are very important, and the contributions from infrared radiation must be considered to obtain a complete energy budget (Wesely and Lipschutz, 1976).

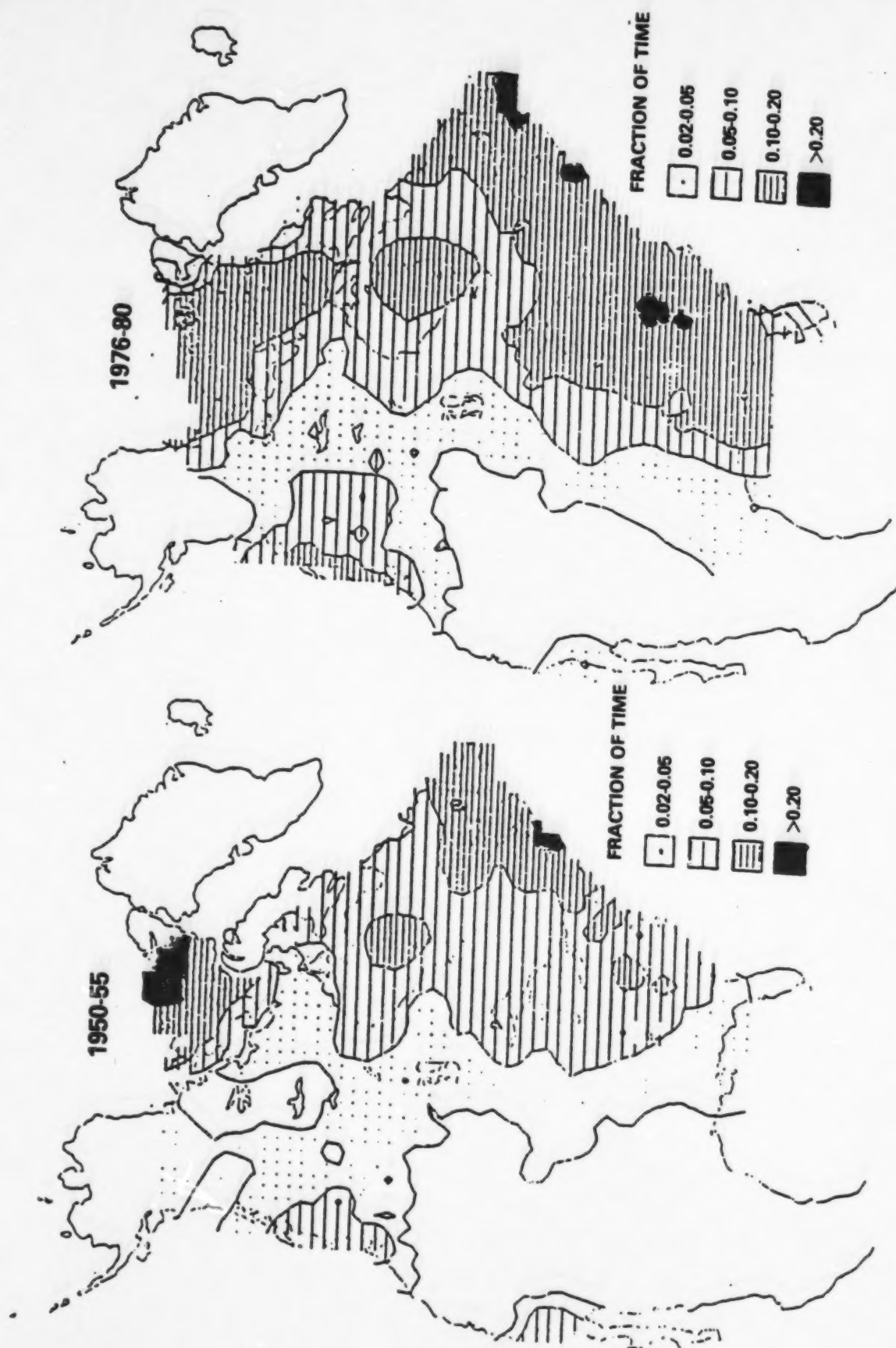


Figure 22. Fog observations for summer seasons.
(3rd quarter)

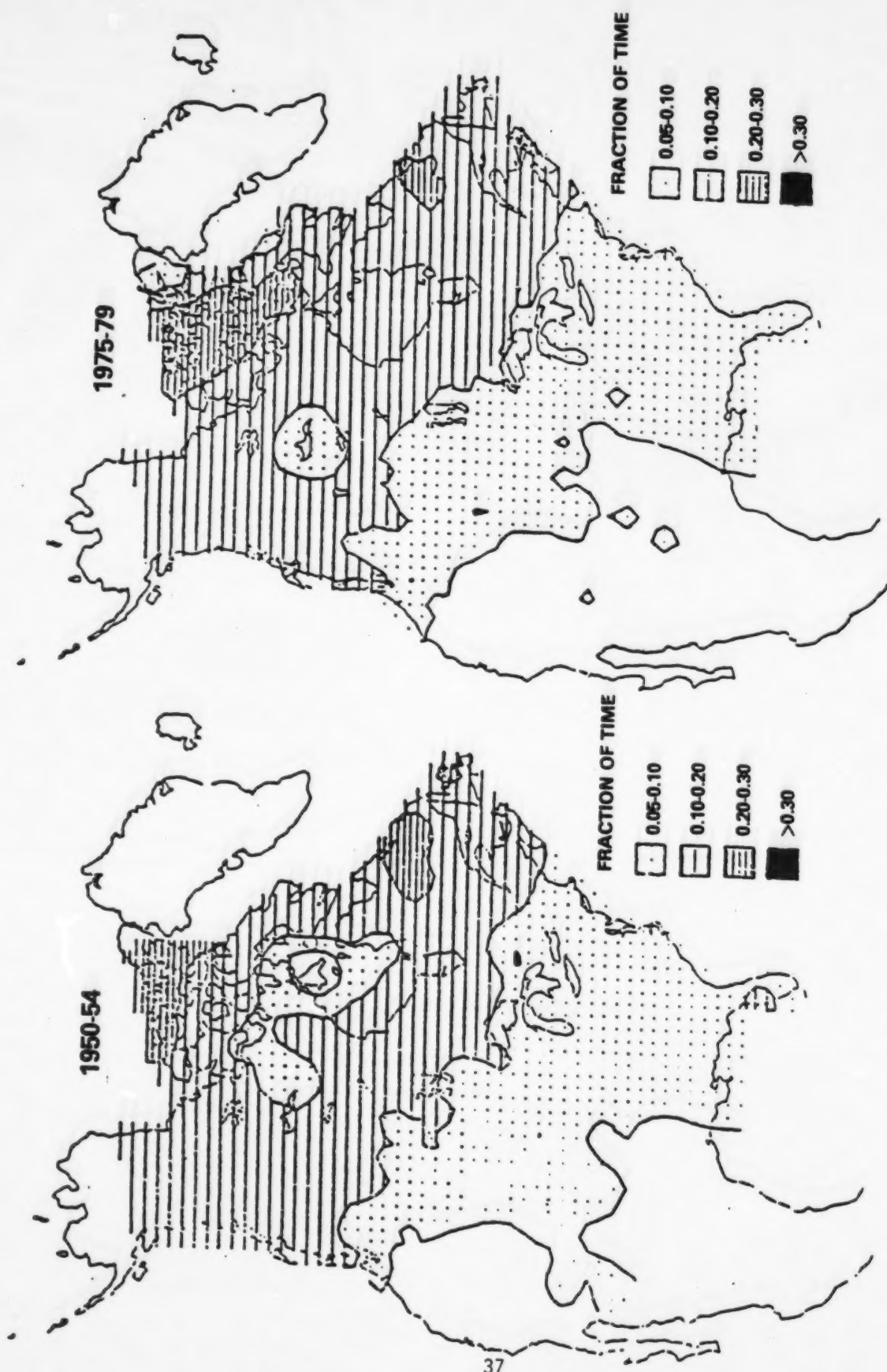


Figure 23. Fraction of time with precipitation for summer season.
(3rd quarter)

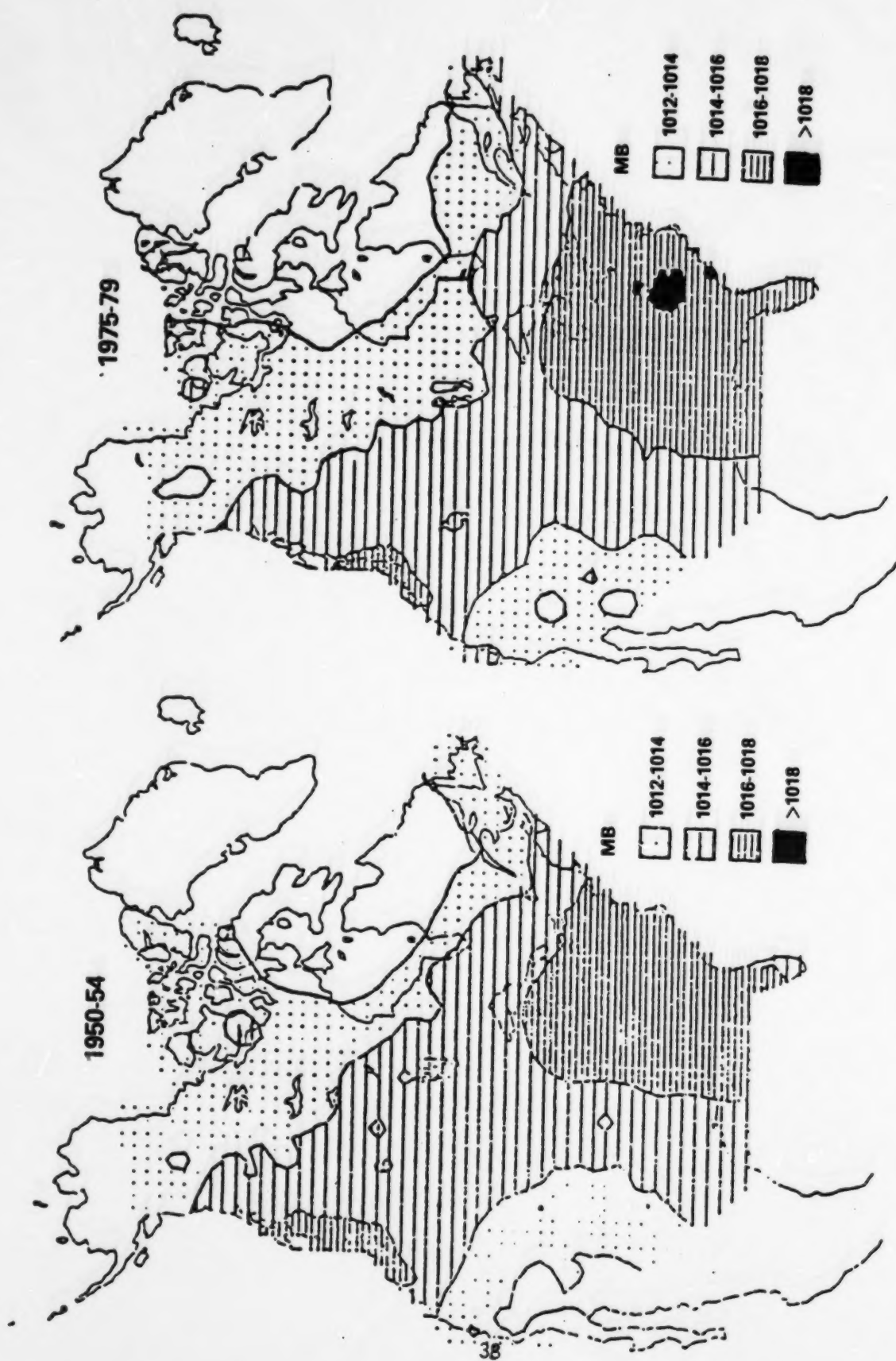


Figure 24. Noon sea level pressure field for summer season.
(3rd quarter)

The importance of particulate matter on climate may be overshadowed by that of CO₂. A doubling of the concentration of CO₂ could result in a 1.5° to 3° C warming of the lower atmosphere, according to various predictions reviewed by the U.S. Department of Energy (U.S. Department of Energy, 1978). One set of calculations frequently cited in the scientific literature predicts a 2° to 3° C rise in the average temperature of the lower atmosphere in the middle latitudes with the doubling of the CO₂ content of the air and a threefold to fourfold greater temperature increase in the polar regions (Manabe and Wetherald, 1975). Current model estimates suggest that the earth should have experienced a few tenths of a degree of warming since the late 1880s due to the increase of CO₂ concentrations from about 290 to the current 335 ppm. However, it appears either that natural variations currently are large enough to mask the expected CO₂ effect on temperature (U.S. Department of Energy, 1980; and Stuiver, 1980) or that models overpredict the magnitude of the effect.

The influence of particles on weather and climate is a complex combination of direct effects on solar and infrared radiation, clouds, and possible synoptic weather patterns. Of these effects, only the direct influence of submicrometer particles on solar radiation can presently be quantified. Aerosols are most likely the principal agents by which pollutants may affect weather and climate, by influencing the structure and distribution of clouds.

The local climatic consequences of the haze of fine particles in the atmosphere are complex and call for sophisticated methods of investigation. Assessment of the effects of the changes on human activity introduces a new level of complexity.

Although it is not possible to calculate accurately the influence the loss of radiation due to scattering and absorption might have on surface temperature, rate of thawing of frozen ground, growing season, or other climatological measures, it is highly probable that this loss cools the ground and heats the hazy lower layers of the atmosphere. In turn, it must increase atmospheric stability, decrease convective mixing, and therefore increase the rate at which pollutants accumulate.

It is possible that local and regional-scale changes in solar radiation caused by human activity may ultimately influence the heat and water vapor content of the atmosphere on very large scales, but solar radiation and aerosol levels measured at stations remote from pollutant sources have not as yet displayed any trend that can be related to human causes (Fischer, 1967; Ellis and Pueschel, 1971; Hodge et al., 1972).

Yet if the effects in industrial regions are strong enough, then the heat balance of the entire earth could be influenced. Additionally, effects from long-lived aerosols, such as those in the stratosphere, might lead to direct physical effects on a global scale.

Regarding the possible climatological impact of sulfur in the troposphere, computations indicate that the change of scattered solar radiation in the polluted areas of the eastern U.S. and western Europe today might well correspond to a temperature drop of several degrees centigrade, though in reality the effect is less, due to ventilation of these areas by general atmospheric circulation.

It should be clear that we are approaching a time when the magnitude of indirect effects of anthropogenic sources may be comparable to natural changes of the climate over centuries. However, the large variability of weather and the self-preserving aspects of the atmospheric system tend to obscure all but the most dramatic changes in climate.

V. Economic Effects.

It is clear that visibility deterioration has economic costs (Rowe and Chestnut, 1982). However, the degree of severity and significance of these costs remain unclear. With society's increasing affluence, tourism has become a major industry. Thus, the impact of aesthetic deterioration due to reductions in visual range is an important issue. Surveys indicate a depth of public concern over such considerations. In some areas, there is also concern over the impact of haze on residential property values.

This visibility problem has been recognized in the U.S., and the National Clean Air Act establishes the requirement that existing standards be maintained or improved, particularly in certain so-called Class I areas (e.g., national parks).

Hazards to ground and air transportation have been associated with greatly reduced visibility caused by high concentration of fine particulate matter. Although the effects on ground transportation from incidents of reduced visibility owing to air pollution are not well documented, they are probably minimal. On the other hand, the effects on aircraft operations are both well documented and sometimes significant. At low visibility, noninstrumental pilots or planes can be grounded and commercial air traffic operations at major airports may be reduced (U.S. Environmental Protection Agency, 1982).

Extra fuel can be required if airplanes are held farther apart for longer times in landing patterns.

Since the 1960's, atmospheric transparency (or turbidity) has also been an important factor in determining the transmission of reflected or emitted radiation reaching remote sensing satellites. With our increasing dependence on such devices, this could well become an area of major concern. The performance of these high technology devices is hampered by unpredictable atmospheric effects much more than by the sensitivity of the detector systems. The implications are that the spectrum range of interest will be much expanded beyond the visible range, particularly into the infrared.

Two areas of concern center on energy. One is the question of how increased absorption and scattering of solar energy will affect the commercial exploitation of this energy source. How will the development of solar energy technologies be affected? The other relates to how alterations in local or regional temperatures affect energy consumption.

Another area of concern is that of impact on biological productivity. Direct effects of aerosols include an increase of the diffuse to direct solar radiation ratio, lowered ground temperature, and shortened daily light doses. Indirect effects include regional climatic changes in quantities such as ground and air temperature, relative humidity, cloud amount, and precipitation. Radiation measurements and modeling indicate that man-caused aerosols reduce the available solar energy from 7 to 20%. Episodic occurrences of regional "haze blobs" that occur during spring, summer, and fall can double or triple these figures. This very large reduction in energy supply has some possible significant effects on biological productivity, (agronomic and forest crops).

The most obvious possibility for influence on crop growth is the reduction in the potential for photosynthesis accompanying reduced solar irradiance. This does not appear to be a significant factor within the U.S. where the available irradiance is far from being a limiting factor on growth. In a higher latitude industrial area (e.g., some northern European regions), the loss of light may be significant.

A second possibility is a reduction of the growing season following reduction of soil temperature in the immediate sub-surface soil layers in spring and fall. Computations have shown that haze can delay the attainment of a given level of irradiance by about one week in March and early April. The loss of 7 to 10 days in planting is comparable to moving 100 miles north. For example, this may represent a 4 to 6% loss in soybean yield.

However, for many of the crops grown in the U.S., heat stress and water supply in the summer months produce a more serious limitation on growth than the dates of germination and first frost. Haze situations can significantly reduce evaporation, which might well be advantageous from the point of view of the yield of some crops in some regions. A reliable assessment of the effects of the haze on crop yield appears to call for the use of a complicated plant growth model for the full growth period. Thus, precise effects on agricultural productivity are still largely a matter of speculation.

Knoerr et al. (1980) discussed the potential impact of decreased solar radiation (due to atmospheric haze) on biological productivity. While solar radiation is related to biological productivity, this relationship is complex. Photosynthesis in some plants becomes light-saturated below maximum levels of solar radiation. For these species some reduction in solar radiation may have essentially no impact on primary production.

However, both photosynthesis and respiration, the net balance of which determines primary production, are also temperature-dependent. Thus, there is an indirect effect of changes in solar radiation on primary production through changes in ambient temperatures. Primary productivity is also dependent upon an adequate supply of soil moisture. Thus, potential changes in primary production, from changes in solar radiation, may be either enhanced or counteracted if there are associated changes in the regional climatic patterns of precipitation and temperature.

A reduction in surface energy supply also reduces the energy-using processes of evapotranspiration, surface air heating, and soil heating. Resultant surface air and soil temperature reduction may also have a direct effect on agricultural and forest production. For many plants, biological production is directly proportional to the sum of mean daily temperature (above some threshold value) during the growing season. Thus, even small reductions in daily temperature values, accumulated over the growing season, have the potential for significant reductions in crop production.

The U.S. Department of Agriculture and the Environmental Protection Agency have been working together on assessments of the economic consequences of alternative ambient ozone levels on agriculture, including effects on producer and consumer well-being (Heck et al., 1983). These models are prototypes for assessing the impact of a number of pollutants in a combined, systemic cost-benefit analysis. Initial interest in this area in 1978 was motivated by

the need to improve on the incorrect or inappropriate procedures used in past assessments of the benefits of pollution control to agriculture.

The need for crop response data to support secondary National Ambient Air Quality Standards and to provide input into economic assessments motivated the Environmental Protection Agency to establish the National Crop Loss Assessment Network (NCLAN). One major determinant of the utility of dose-response information is the degree to which it contributes to improving the economic efficiency basis of environmental policy decisions (Adams and Crocker, 1982). In order to improve economic assessments, NCLAN funded a series of regional and preliminary national economic assessments in 1982 and 1983 intended to test methodologies, define data needs and provide benefits estimates for alternative ozone standards.

The research thrust in the NCLAN and peripheral research projects has been on agricultural crops. While the economic consequences of environmental change on agriculture appear substantial, the magnitude of these effects vis à vis other welfare categories (e.g., materials, other vegetation values) is not well established.

Capturing the economic effects of environmental change on nonmanaged ecosystems is conceptually and empirically more difficult and hence few economic assessments of these phenomena exist. Some of these effects are to be addressed in NCLAN in 1986 as the emphasis shifts from crops to forests.

The policy utility of the few studies in the literature is limited to specific costs and hence is insufficient to gauge either directions or magnitude of probable economic effects on the national level. Assessing and integrating these diverse economic effects into a total benefit-cost analysis poses major challenges and perhaps some of the greatest policy payoffs for environmental economics.

REFERENCES

- Abbot, C. J. (1911) *The Sun*, Appleton, N.Y.
- Adams, R. M.; Crocker, T. D. (1982) Dose response information and environmental damage assessment: An economic perspective. *J. Air Pollut. Control Assoc.* 32: 1062-1067.
- Altshuller, A. P. (1980) Seasonal and episodic trends in sulfate concentration (1963-1978) in the eastern United States. *Environ. Sci. Technol.* 14: 1337-1349.
- Angstrom, A. (1930) On the atmospheric transmission of sun radiation. II. *Geografiska Annaler* 12: 139-159.
- Atwater, M. A. (1970) Planetary albedo changes due to aerosols. *Science* (Washington, D.C.) 170: 164-166.
- Ball, R. J.; Robinson, G. D. (1982) The origin of haze in the Central United States and its effects on solar irradiation. *J. Appl. Meteorol.* 21: 171-188.
- Breslau, N.; Dave, J. V. (1973) Effects of aerosols on the transfer of solar energy through realistic model atmospheres. Part I. Nonabsorbing aerosols. *J. Appl. Meteorol.* 12: 601-615.
- Bryson, R. A. (1968) All other factors being constant. *Weatherwise* 21: 56-61.
- Carlson, T. (1984) Comments, summarized in Summary of the Workshop on the Effects of Air Pollutants on Climate and Visibility. Quail Roost Conference Center, Rougemont, N.C.: U.S. Environmental Protection Agency, January 29-February 1.
- Charlson, R. J.; Robinson, G. D. (1984) The influence of fine particles on local and regional scale radiative climate. Presented at: Workshop on the Effects of Air Pollutants on Climate and Visibility. Quail Roost Conference Center, Rougemont, N.C.: U.S. Environmental Protection Agency, Environmental Sciences Research Laboratory; January 29-February 1. Available at Environmental Criteria and Assessment Office, Research Triangle Park, NC 27711.
- Charlson, R. J.; Pilat, M. J. (1969) Climate the influence of aerosols. *J. Appl. Meteorol.* 8: 1001-1002.
- Charlson, R. J.; Vanderpol, A. H.; Covert, D. S.; Waggoner, A. P.; Ahlquist, N. C. (1974) Sulfuric acid-ammonium sulfate aerosol: Optical detection in the St. Louis region. *Science* (Washington, D.C.) 184: 156-158.
- Coakley, J. A.; Schneider, S. H.; Dickinson, R. E. (1975) Computation of possible climatic consequences of perturbations to stratospheric aerosols, ozone and water vapor with radiative-convective models. Final report. Boulder, CO: National Center for Atmospheric Research.

- Dzubay, T. G.; Stevens, R. K.; Lewis, C. W. (1982) Visibility and aerosol composition in Houston, Texas. *Environ. Sci. Technol.* 16: 514-525.
- Ellis, H. T.; Pueschel, R. F. (1971) Solar radiation: Absence of air pollution trends at Mauna Loa. *Science* (Washington, D.C.) 172: 845-846.
- Ensor, D. S.; Porch, W. M.; Pilat, M. J.; Charlson, R. J. (1971) Influence of the atmospheric aerosol on albedo. *J. Appl. Meteorol.* 10: 1303-1306.
- Fischer, W. H. (1967) Some atmospheric turbidity measurements in the Antarctic. *J. Appl. Meteorol.* 6: 958-962.
- Flowers, E. C.; McCormick, R. A.; Kurfis, K. R. (1969) Atmospheric turbidity over the United States, 1961-1966. *J. Appl. Meteorol.* 8: 955-962.
- Heck, W. W.; Adams, R. M.; Cure, W. W.; Heagle, A. S.; Heggstad, H. E.; Kohut, R. J.; Kress, L. W.; Rawlings, J. O.; Taylor, O. C. (1983) A reassessment of crop loss from ozone. *Environ. Sci. Technol.* 17: 573A-581A.
- Hobbs, P. V.; Harrison, H.; Robinson, E. (1974) Atmospheric effects of pollutants. *Science* (Washington, D.C.) 183: 909-915.
- Hodge, P. V.; Harrison, H.; Robinson, E. (1972) Astronomy and air pollution. *Science* (Washington, D.C.) 178: 1123-1124.
- Husar, R. B.; Patterson, D. E.; Holloway, J. M.; Wilson, W. E.; Ellestad, T. G. (1979) Trends of eastern United States haziness since 1948. In: *Proceedings of the fourth symposium on atmospheric turbulence, diffusion, and air pollution*, American Meteorological Society; Reno, NE. Pp. 249-256.
- Knoerr, K. R.; Reynolds, J. F.; Strain, B. R.; Cunningham, G. L. (1980) Predicting primary productivity for forest and desert ecosystem models. In: Hesketh, J. D.; Jones, J. W., eds. *Predicting photosynthesis for ecosystem models: V. II*. Cleveland, OH: CRC Press; pp. 169-207.
- Liken, F.; Boda, K. (1922) Vorschagezur Berechnung des Trubungsgrades der atmosphere aus messungen der sonnenstrahlung. *Meteor. Z.* 39: 161.
- Lin, C. I.; Baker, M.; Charlson, R. J. (1973) Absorption coefficient of atmospheric aerosol: A method for measurement. *Appl. Opt.* 12: 1356-1363.
- Machta, L.; Telegadas, K. (1974) Inadvertent large-scale weather modification. In: Hess, W. N., ed. *Weather and climate modification*. New York, NY: John Wiley and Sons; pp. 687-725.
- Macias, E. S.; Zwicker, J. O.; Ouimette, J. R.; Hering, S. V.; Friedlander, S. K.; Cahill, T. A.; Kuhlmeier, G. A.; Richards, L. W. (1981a) Regional haze case studies in the southwestern U.S. I. Aerosol chemical composition. In: *Proceedings of the symposium on plumes and visibility. Measurements and model components*; November 1980; Grand Canyon National Park, AZ. *Atmos. Environ.* 15: 1971-1986.

- Macias, E. S.; Zwicker, J. O.; White, W. H. (1981b) Regional haze case studies in the southwestern U.S. - II. Source contributions. In: Proceedings of the symposium on plumes and visibility. Measurements and model components; November 1980; Grand Canyon National Park, AZ. Atmos. Environ. 15: 1987-1997.
- Manabe, F.; Wetherald, R. (1975) The effects of doubling the CO₂ concentration on the climate of a general circulation model. J. Atmos. Sci. 32: 3-15.
- McCormick, R. A.; Ludwig, J. H. (1967) Climate modification by atmospheric aerosols. Science (Washington, D.C.) 156: 1358-1359.
- Mitchell, J. M. (1971) The effects of atmospheric aerosols on climate with special reference to temperature near the earth's surface. J. Appl. Meteorol. 10: 703-714.
- National Research Council. (1983) Changing climate. Washington, DC: National Academy Press.
- Reck, R. A. (1975) Aerosols in the atmosphere: Calculation of the critical absorption/backscatter ratio. Science (Washington, D.C.) 186: 1034-1036.
- Reck, R. A. (1975) Aerosols and polar temperature changes. Science (Washington, D.C.) 188: 728-730.
- Reck, R. A. (1977) Influence of airborne particles on the earth's radiation balance. Warren, MI: General Motors Research Laboratories; report GMR 2565.
- Reitan, C. H. (1960) Mean monthly values of precipitable water over the United States, 1946-1956. Mon. Weather Rev. 88: 25-35.
- Robinson, G. D. (1977) Effluents of energy production: Particulates. In: Energy and Climate, Studies in Geophysics, Geophysics Study Committee, Research Board, National Academy of Sciences; pp. 61-71.
- Rowe, R. D.; Chestnut, L. G. (1982) The value of visibility: Economic theory and applications for air pollution control. Cambridge, MA: Abt Books.
- Stuiver, M. (1980) Atmospheric carbon dioxide and carbon reservoir changes. Science (Washington, D.C.) 199: 253-258.
- U.S.C. (1977) 42: § 7491. Clean Air Act, § 169A, visibility protection for Federal class I areas.
- U.S. Department of Energy, Office of Carbon Dioxide Effects Research and Assessment. (1980) A comprehensive plan for carbon dioxide effects, research, and assessment. Part I: The global carbon dioxide effects of increasing carbon dioxide. Washington, D.C.: U.S Department of Energy; DOE/EV-0094.
- Waggoner, A. P.; Vanderpol, A. J.; Charlson, R. J.; Larsen, S.; Grant, L.; Tragardh, C. (1976) Sulphate-light scattering ratio as an index of the role of sulphur in tropospheric optics. Nature (London) 261: 120-122.

- Waggoner, A. P.; Weiss, R. E.; Ahlquist, N. C.; Covert, D. S.; Will, S.; Charlson, R. J. (1981) Optical characteristics of atmospheric aerosols. Atmos. Environ. 15: 1891-1909.
- Waggoner, A. P.; Weiss, R. E.; Larson, I. V. (1982) In situ rapid response measurement of $\text{H}_2\text{SO}_4/(\text{NH}_4)_2\text{SO}_4$ aerosols in urban Houston: a comparison with rural Virginia. Submitted to Atmospheric Environment, August.
- Weiss, R. E. (1980) The optical absorption properties of suspended particles in the lower troposphere at visible wavelengths. Seattle, WA: University of Washington; Ph.D. Thesis. Available from University Microfilms, Ann Arbor, MI.
- Weiss, R. E.; Waggoner, A. P.; Charlson, R. J.; Ahlquist, N. C. (1977) Sulfate aerosol: Its geographical extent in the midwestern and southern United States. Science (Washington, D.C.) 195: 979-981.
- Weseley, M. L.; Lipschutz, R. C. (1976) An experimental study of the effects of aerosols on diffuse and direct solar radiation received during the summer near Chicago. Atmos. Environ. 10: 981-987.
- WMO/EPA/NOAA/UNEP. (1974-1977) Global monitoring of the environment for selected atmospheric constituents, 1972-1975. Asheville, NC: Environmental Data Service National Climatic Center.